Gap formation in the density of states as the indicator of metal-insulator transition caused by electron-phonon coupling

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Abstract. This research explores the electron-phonon coupling effect on manganite system transport properties. Using the Holstein Phonon model, which has been documented to capture metal-insulator transitions in single band model, we investigate the electron-phonon coupling effect. We also look into insulator phase forming criteria, using more realistic phonon model for manganite system; the Jahn-Teller model. To observe the forming of gap energy in Jahn-Teller model, we also do the separate computation for Q2 and Q3 modes. For this computation, interactions between electron spins and Mn ions spin are omitted. Computation is done using Dynamical Mean Field Theory (DMFT). The computation results are shown through the Density of State (DoS) Profile for various modes, electron filling, electron-phonon coupling, and temperature. Our computation results prove that Jahn-Teller model with a certain minimum electron-phonon coupling constant (g) can produce energy gap, where chemical potential (µ) fall precisely in the middle of energy gap that causes the system to be an insulator. This result applies to any electron filling value (x). Computation with electron-phonon coupling variation and temperature shows that the width of the energy gap increases along with the increase of both variables.

1. Introduction

MIT (Metal-insulator transition) is a phenomenon in which a certain material experience transformation of electrical property from a metallic phase to insulator phase or vise versa. Such phenomenon first realized by Boer and Verwey when they observe several kinds of metal oxide transition like NiO which band energy of $d$ orbital occupied half-filling that define insulator property [1]. Oppositely, band, energy theory said such condition should define metal property [2]. So that there were obvious observation and researches conducted to explain how the mechanism of that transition can occur, moreover when MIT phenomenon discovered in electronic device [3]. In 1937, Mott and Pierls suggested that Coulomb interaction among electrons or electrons correlation might be caused MIT phenomenon [4]. According to their suggestion, electrons correlation very likely lead the electrons to be localized so that modification of electron’s theory in metal were needed by adding such interaction. From here the materials which correspond with MIT phenomenon called Mott insulator [5].

In the growth of study, there were several mechanisms proposed to explain MIT phenomenon. Pierls proposed electron-phonon interaction because of crystal structure deformation [6]. Anderson proposed randomness of crystal structure [7]. Slater proposed magnetic moment alignment [8]. The
classification of materials whether having Mott transition, Pierls, Anderson or Slater has been done. In contrast to materials that can be identified precisely its electronic properties, systems which close to MIT can undergo phase change because of physical properties change dramatically with the variation of control parameters such as carrier concentration, temperature, interaction, or external magnetic field [9]. There are several models which can describe theoretically strongly correlated electron systems especially Mot transition such as the Hubbard model[10] and Anderson impurity model [11].

Mixed valence manganese oxides $R_1xA_xMn_{3-x}O_y$, we define R is a trivalent rare-earth, and A is a divalent alkaline earth ion. This material has been studied since 1950, especially for one type of manganese oxide ie manganiteperovskites. First examined by Jonker and Van Santen[12]. They found the antiferromagnetic isolator properties in the doping region of x such as $x = 0$ and $x = 1$. At $x = 3$ they found a ferromagnetic change associated with a sharp increase of electrical conductivity or so-called paramagnetic transition of insulator-ferromagnetic metal (PI-FM). In 1951 Zener proposed a double-exchange model to explain the mechanism of this PI-FM [13]. This calculation fits qualitatively with manganites experiments. But when compared to experimentally obtained transition temperatures the results of the double-exchange model are well above the experiments. Then in 1995 Millis proposed a calculation that takes into account the influence of the Jahn-Teller phonon [14]. By incorporating the phonon's influence, the new model can explain the phenomenon of PI-FM transition in LCMO for doping $x = 0.3$. In 2007 Ramakhrisnan also proposed a similar calculation of the Jahn-Teller phonon model with the addition of Hubbard interaction (onsite-repulsion) and produced a calculation that could show the PI-FM transition phenomenon in LCMO [15].

The latest experimental results from Rusydiet al show the optical conductivity curve for LCMO and explain the presence of doping and temperature dependence on the high energy photon spectrum [15]. The calculation by Millis and Ramakhiran are not appropriate to explain the results of Rusydi et al. experiments as these two models do not apply to electron systems in high energy spectrum where optical conductivity occurs. One model that attempts to explain the experimental results of Rusydiet al is the Majidiet al calculation in which proposes the role of oxygen atoms in the calculations [16]. The calculation proposed by Majidiet al. succeeds in explaining the experimental results of Rusydi et al but not for the low energy spectrum electron system region where there is a PI-FM transition on the energy spectrum 0 eV. As for the low energy spectrum, Majidiet al. suggest that the PI-FM transition phenomenon may occur because of attenuation and strengthening of MnO parameter hopping triggered by spin-spin interaction and electron-phonon coupling [16].

We conduct this study to see the correspondence between electron-phonon interaction with the formation of the insulating phase in the visualization of a density of states. As for the formulation for electron-phonon coupling interactions we use two models namely the Holstein model and Jahn-Teller model. First, we work with the Holstein model because of its simplest formulation, and then we do with Jahn-Teller model because it is more realistic and has not implemented before[17]. Using our manganite model base, we hope to be able to explain the general MIT phenomenon through criteria of coupling, impurity, and temperature parameters.

2. Methods

2.1. Hamiltonian

According to Jahn-Teller distortion theory, Mn 3d orbital degenerated to five folding there are two kinds of $e_g$ orbital $(d_{xy}, d_{xz}, d_{yz})$ and three kinds of $t_{2g}$ orbital $e_g (d_{xy}, d_{yz}, d_{xy})$ [18]. Contrast to $e_g$ orbital which has a higher energy state, $t_{2g}$ orbital tightly bound to the core spin with spin's number $S = \frac{3}{2}$. Here we propose the Hamiltonian based on Hilbert space which constructed from 4 basis $e_g$ orbital $\{\text{Mn}-e_g x^2 - y^2, \uparrow\}$, $\{\text{Mn}-e_g x^2 - y^2, \downarrow\}$, $\{\text{Mn}-e_g 3z^2 - r^2, \uparrow\}$, $\{\text{Mn}-e_g 3z^2 - r^2, \downarrow\}$. The upper and lower states are associated with Jahn-Teller splitting.

$$H = \sum_{k, \sigma} \epsilon(k) c_{k, \sigma}^\dagger c_{k, \sigma} + \sum_{i} \left( \frac{\mu_1}{2M} + \frac{1}{2} M \omega^2 x_i^2 \right) + g \sum_{\tau} (Q_{21} \tau_1^x + Q_{31} \tau_3^z)$$

(1)

The following is a Hamiltonian formulation that we use to perform calculations. In the first term of Hamiltonian describes onsite energy with $\epsilon(k)$ is kinetic energy spectrum of the electron, $c_{k, \sigma}^\dagger$ and $c_{k, \sigma}$
are the creation and annihilation operators for electrons on-site \( i \) within \( e_g \) orbital and spin \( \sigma \). The second term describes Jahn-Teller kinetic energy associated to Hook’s law. The third term describes local Jahn-Teller interaction where \( g \) is Jahn-Teller coupling, \( Q_{z1} \) and \( Q_{z3} \) are the Jahn-Teller displacements associated with two different vibrations of Mn ion, \( \tau_{x,y,z}^{i} = \sum_{a} \beta_{a}^{\dagger} \sigma_{a}^{x,y,z} \beta_{a} \), where \( \sigma_{a}^{x,y,z} \) is Pauli matrices [17]. As a note when we perform the calculation in Holstein phonon model framework, we turn off the \( Q_{z1} \) so that the formulation becomes equal to \( \sum_{i} n_{i} x_{i} \) where \( n \) is a density of i-site and \( x \) is displacement.

2.2. Dynamical Mean Field Theory

The basic concept of DMFT calculation is employing the Green function[19].

\[
\begin{align*}
[G(k, \omega)] &= \{[G(\omega)]_{\text{eff}} - [\Sigma(\omega)]\}^{-1} \\
G_{mf}(x) &= \{[G(\omega)]_{\text{eff}} - [\Sigma(\omega)]\}^{-1}
\end{align*}
\]

Steps DMFT method is as follows. First of all, coarse-graining on the Green function or averaged around Brillouin zones to eliminate dependence on momentum so that Green function is localized (not depend on position).

\[
G(\omega) = \frac{1}{N} \sum_{k} [G(k, \omega)]
\]

From the Green mean field function, then we look for the local Green function \( G_{loc}(\omega, Q_{2}, Q_{3}) \).

Here we apply Hamiltonian of electron-phonon interaction to obtain local interaction \( \Sigma_{loc} \).

\[
\Sigma_{loc}(\omega, Q_{2}, Q_{3}) = \left\{ \begin{array}{ccc}
g_{Q_{3}} & g_{Q_{2}} & 0 \\
g_{Q_{2}} & -g_{Q_{3}} & 0 \\
0 & g_{Q_{3}} & g_{Q_{3}} \\
0 & g_{Q_{2}} & -g_{Q_{3}} \end{array} \right\}
\]

Then we look for probability \( P(x, \theta) \) of each spin \( S \) on each site by counting the effective action \( S(x, \theta) \) and local partition function \( Z \) [19].

\[
P(x, \theta) = \frac{e^{-S(x, \theta)}}{Z}
\]

\[
S(x, \theta) = \frac{k(Q_{x}^{2} + Q_{y}^{2})}{2k_B T} + \frac{H_S \cos \theta}{k_B T} - \sum_{n} \ln \det[G_{loc}(i \omega, Q_{2}, Q_{3})] e^{-i \omega \theta}
\]

\[
Z = \int \frac{e^{-S(Q_{2}, Q_{3}, \theta)}}{\sin \theta} dx \, d\theta
\]

Average Green function \( G_{ave}(\omega) \) obtained by:

\[
G_{ave}(\omega) = \int P(x, \theta) G_{loc}(x, \theta) \, d\theta
\]

With the gain of \( G_{mf}(\omega) \) and \( G_{ave}(\omega) \) we recalculate self-energy \( \Sigma(\omega) \) as following :

\[
\Sigma(\omega) = \left[ G_{mf}(\omega) \right]^{-1} - [G_{ave}(\omega)]^{-1}
\]

The iteration process is followed by a real frequency value \( (\omega = \omega + i \delta) \) so there will be a convergent self-energy value. After getting the convergent calculation, the value of DOS (density of state) is obtained by the equation:

\[
\text{DOS}(\varepsilon) = -\frac{1}{\pi} \text{Im} \text{Tr} \left[ G_{ave, R}(\varepsilon) \right]
\]

With \( R \) expressing the real frequency. Then potential chemical value inside the doping state of \( x \) will be obtained by the equation:

\[
b = (1 - x)N = \int F(\varepsilon) \text{DOS}(\varepsilon) d\varepsilon
\]

\[
f(\varepsilon) = \frac{1}{e^{\frac{\varepsilon}{k_B T}} + 1}
\]
With \( b \) is the doping density of the non-doping particles, \( x \) is doping, \( N \) is the sum of basis, \( f(\varepsilon) \) is a Fermi-Dirac distribution function, \( \mu \) as chemical potential, \( \varepsilon \) is energy, and \( T \) is temperature. By knowing DOS parameters and chemical potentials, we can see whether the system is in an insulator or conductor state.

3. Result and Discussion

Previous Holstein model calculations have shown the effect of interaction electron-phonon that affect gap formation in the density of states\[20\]. Here we will show the modification from a previous calculation by adding the basis state to double band that previously only used a single band. Use the electron \( x \) filling variations we used are 0.1, 0.3, 0.5, 0.7, and 0.9 with \( x \) is a rare earth atom such as Ca. We need to convey as a comparison that on Hubard's model can only guarantee the occurrence of metal-insulator transitions on the circumstances charging half-filling, while for a sufficient massive \( x \) value (away from half filling) Hubard model calculation has not been able to guarantee the occurrence transition metal-insulator\[10\]. Effective electron-phonon coupling parameters \( (g) \) are effective to generate a gap on the Holstein model is 0.7, in contrast to the parameters the clutch for the Jahn-Teller model is 0.9. The DOS results in Figure 1.a show the formation of energy gaps in which the chemical potential takes place. The chemical potential that falls right in the gap indicates that in the middle of the gap is a state that can not be occupied by electrons, in other words, has formed insulator phase.

![Figure 1(a)](image1.png)

![Figure 1(b)](image2.png)

**Figure 1.(a)** DOS for Holstein mode system by filling electrons at temperature 300 K, (b) DOS for Jahn-Teller mode system with variations of electron filling in temperature 300 K.

The results of Millis’s calculation in phonon vibration being one of the causes of PI-FM transition in LCMO by inhibiting the movement of electrons and making the electron isolated on a particular site\[14\]. The vibration mode that contributes to this PI-FM transition is known as Jahn-Teller phonon.

After working on a Holstein model that can ensure the transition metal-insulator, in Fig.1.b we show the results of Jahn-Teller's model also able to guarantee the transition metal-insulator. In calculations, we divide this Jahn-Teller model into three modes namely Jahn-Teller mode, Q2 mode, and Q3 modes. The Jahn-Teller vibration model accommodates the system of two vibration orientation directions; Q2 and Q3. When Q2 and Q3 actively oscillate with a coupling parameter \( (g) \), then this is what we call Jahn-Teller mode. Q2 mode is when we switch off Q3 and vice versa. Figure 4.2 shows the variations in electrons that produce \( x \) pseudogap and the exact chemical potential value falls in the middle of the pseudogap. Value the opposite of the Holstein model also occurs for the Jahn-Teller model, but the DOS curve for electron was filling \( x = 0.1 \) coincides with DOS for charging electron \( s \) \( x = 0.9 \), as well as DOS for electron filling \( x = 0.3 \) and \( x = 0.7 \). While the pseudogap that is formed cannot be said gap which shows the system in a state of the insulator phase, but we believe if the electron-phonon coupling parameter value \( (g) \) we magnify the pseudogap will turn into a gap like we did in Fig.2. As for the real gap is obtained by the value of electron charging \( x = 0.5 \) where the value is gap is almost worth 2 eV. We suspect the electron filling value \( x = 0.5 \) is the value the optimal
electron filling that can produce the gap, it is highly relevant to DOS peaks that initially are in the middle is slowly declining when the electron was filling value approaches $x = 0.5$, either approximated from the lower limit or the upper limit.

![Figure 2](image1.png)  
**Figure 2.** (c) DOS for Q2 mode system with variations of electron filling at temperature = 300 K, (d) DOS for Q3 mode system with variations of electron filling at temperature = 300 K.

![Figure 3](image2.png)  
**Figure 3.** (e) DOS for Jahn-Teller mode with variations of coupling parameter value ($g$) at temperature = 300 K, (f) DOS for Jahn-Teller mode with temperature variations.

To know the dependence of DOS on electron-phonon coupling parameters and temperature, we display the Fig.3.e and Fig.3.f. The coupling value starts to form pseudogap at the time value $g = 0.7$, while for value $g = 0.9$ has formed a pseudogap which in almost touching the limit DOS = 0 so that we make $g = 0.9$ is the critical value of the electron-phonon coupling parameter. The value of the coupling parameter $g$ which is already effective produces a real gap in the Fig.3.e is $g = 1$ which is blue. If the coupling parameter value $g$ continues to be enlarged, then the energy gap or gap will continue to widen. DOS curve as a function of temperature indicates that the effect of increasing temperatures can gradually narrow the gap, even when the temperature modulated to 400 K, the gap has changed to pseudogap. Also the effect of temperature rise also causes DOS to be shorter.

**4. Conclusion**

Our calculations confirm that Holstein's model with a certain electron-phonon coupling ($g$) can produce an energy gap, where the chemical potential position ($\mu$) falls right in the middle of the energy gap which causes the system to be an insulator. This result applies to any electron filling value ($x$) because the Holstein model is considered naive to describe phonons in the manganite system, in this study we tested the effect of electron-phonon coupling in the more realistic Jahn-Teller model.
Jahn-Teller Mode is a combination of two kinds of modes (Q2 and Q3). To test the contribution each component in the Jahn-Teller model, we do calculating the contribution of Q2 and Q3 modes separately, and The complete Jahn-Teller mode is a combination of Q2 and Q3 modes. Calculation results we show that both Q2 and Q3 modes, as well as combine both, with the value of a certain electron-phonon coupling, turns out, too can produce an insulator phase at any value of electron filling. The difference between the Holstein model calculations and the Jahn-Teller modeling in the resulting density of state (DOS) profile. With the Jahn-Teller model has been done DOS calculations with variations electron-phonon coupling, i.e., 0.5, 0.7, 0.9, and 1, at a fixed temperature value 300 K. Our calculation results show that with $g = 0.7$ has formed pseudo gap. With the value of $g$ enlarged "valley" pseudogap increasingly in, until finally, pseudogap becomes gap at $g = 0.9$. With $g$ expanded again into 1 gap increases in width. With the Jahn-Teller model has also done DOS calculation with temperature variations, i.e., 400 K, 300 K, 200 K, and 100 K, at the coupling value fixed electrons ($g = 0.9$). Our calculation results show that the energy gap widens with decreasing temperature.

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