1. Introduction
Owing to global warming and environment issues, it is important to develop new energies to reduce the total amount of CO$_2$ in the atmosphere. Electrical power generated by the mechanical force of wind and ocean tides is an example of such green energy. Additionally, electrical power yielded from solar energy transformation also creates clean energy. Many technologies are devoted to developing materials with high figure of merit (ZT) for potential applications in solid-state thermal devices, which can be utilized to generate electrical power by the temperature bias resulting from either solar heating or joule heating.

Several methods were proposed to realize the enhancement of ZT, one of them is to reduce system dimension. A zero dimension quantum dot (QD) system was predicted to be more pronounced for the enhancement of thermoelectric efficiency in dimension reduction. Nevertheless, these studies have focused on the linear response regime of $\Delta T/T_0 \ll 1$, where $\Delta T$ and $T_0$ are the temperature bias and equilibrium temperature of two side electrodes, respectively. The thermal properties of thermal devices in the nonlinear response regime $\Delta T/T_0 \sim 1$ have important applications, such as in thermal rectifiers and thermal transistors. A thermal rectifier is crucial for heat current storage.

Records of thermal rectification date back as early as 1935, when Starr discovered that copper oxide/copper junctions can display a thermal diode behavior. The thermal rectification effects have been theoretically predicted to occur in the one dimensional phonon junction system. Additionally, the thermal rectification can also be realized by photon carriers through vacuum. Owing to phonon or photon carriers without charges, their applications are restricted in the heat manipulation and heat energy storage. Using charge carriers (electrons or holes), we can manipulate not only heat currents but also charge currents.

Recently, Scheibner et al. employed a single metallic QD junction to measure the electrochemical potential in the linear response regime. Such a small electrochemical potential yielded by temperature bias is not sufficient to demonstrate the thermal rectification effect. Furthermore, the operation temperature is very low for metallic QDs owing to small charging energies compared with the thermal energy ($k_B T_0$). From an application point of view, the semiconductor QD junctions with large interdot Coulomb interactions and energy-level separation within each QD allow the QD thermal rectifiers to be operated at much higher temperature.

The rectification effect of charge currents has been studied in the multiple QD junctions, and the mechanism of thermal rectification is similar to that of charge current rectification. However, there are two derived forces to yield heat currents in the QD junctions. It is not straightforward to reveal the behavior of heat current rectification owing to the strong nonlinear relationship between the temperature bias and the electrochemical potential. This study has revealed that in the system of multiple semiconductor QDs embedded into an amorphous insulator with low heat conductivity connected with the metallic electrodes the thermal rectification effect exists in the nonlinear response regime. The thermal power in the linear and nonlinear regimes is also clarified.

2. Formalism
The Hamiltonian of the metal/quantum dots/metal double barrier junction shown in Fig. 1 is described by the following multi-level Anderson model:

$$H = \sum_{k,\alpha,\beta} \epsilon_k a_{k,\alpha,\beta}^\dagger a_{k,\alpha,\beta} + \sum_{l,\sigma} E_l d_{l,\sigma}^\dagger d_{l,\sigma}$$

$$+ \sum_{l,\sigma} U_l d_{l,\sigma}^\dagger d_{l,-\sigma} d_{l,-\sigma}^\dagger d_{l,\sigma} + \frac{1}{2} \sum_{l,\sigma,j,\sigma'} U_{l,j} d_{l,\sigma}^\dagger d_{j,\sigma'}^\dagger d_{l,-\sigma} d_{j,-\sigma'}$$

$$+ \sum_{k,\alpha,\beta, l,\sigma, j,\sigma'} V_{k,\alpha,\beta} d_{l,\sigma}^\dagger a_{k,\alpha,\beta} + \sum_{k,\alpha,\beta, j,\sigma'} V^*_{k,\beta,\alpha} a_{j,\sigma'}^\dagger d_{j,\sigma},$$

where $a_{k,\alpha,\beta}^\dagger$ (or $a_{k,\alpha,\beta}$) creates (destroys) an electron of momentum $k$ and spin $\sigma$ with energy $\epsilon_k$ in the $\beta$ metallic electrode. $d_{l,\sigma}^\dagger$ (or $d_{l,\sigma}$) creates (destroys) an electron with the ground-state energy $E_l$ of the $l$th QD, $U_l$ and $U_{l,j}$ describe, respectively, the intradot and interdot Coulomb interactions. $V_{k,\alpha,\beta}$ describes the coupling between the band states of electrodes and the QDs. The Hamiltonian of eq. (1) assumes that energy level separations between the ground state and the first excited state within each QD are much larger than intradot Coulomb interactions $U_l$ and thermal temperature $k_B T$. Therefore, there is only one energy level for each QD. Meanwhile, we have ignored the interdot hopping terms due to the high potential barrier separating QDs.

Using Keldysh–Green’s function technique, the charge and heat currents leaving electrodes can be expressed as

$$J_c = -\frac{2e}{\hbar} \sum_l \int \! \! \! d\epsilon \gamma(\epsilon) \text{Im} G_{l,\sigma}(\epsilon) f_{lR}(\epsilon),$$

$$Q = -\frac{2}{\hbar} \sum_l \int \! \! \! d\epsilon \gamma(\epsilon) \text{Im} G_{l,\sigma}(\epsilon)(\epsilon - E_F - e\Delta V) f_{lR}(\epsilon),$$

where $f_{lR}(\epsilon) = f_L(\epsilon) - f_R(\epsilon)$, the transmission factor is given as

$$\frac{f_{lR}(\epsilon)}{f_{lR}(\epsilon) - f_L(\epsilon)}.$$
is the Fermi energy of electrodes.

is the Fermi distribution function for the left (right) electrode. The chemical potential difference between the two electrodes is related to the bias difference \( \mu_L - \mu_R = e\Delta V \). The temperature difference is \( T_L - T_R = \Delta T \). \( E_F \) is the Fermi energy of electrodes. \( \Gamma_{L(R)}(\epsilon) \) and \( \Gamma_{L(R)}(\epsilon) \) denote the tunneling rates from the QDs to the left and right electrodes, respectively. Notations \( e \) and \( h \) denote the electron charge and Planck’s constant, respectively. For simplicity, these tunneling rates will be assumed to be energy and bias independent. Equations (2) and (3) have been employed to study the thermal properties of a single-level QD in the Kondo regime.\(^{22,23}\) Here, our analysis is devoted to the heat current in the Coulomb blockade regime. The expression of retarded Green function is given in refs. 20 and 24:

\[
G_{\sigma,i}(\epsilon) = (1 - N_{\sigma,-}) \sum_{m=1}^{N_{\sigma}} \frac{p_m}{\epsilon - E_l - \Pi_m - i\Gamma_l} + N_{\sigma,-} \sum_{m=1}^{N_{\sigma}} \frac{p_m}{\epsilon - E_l - \Pi_m + i\Gamma_l},
\]

where \( n \) denotes the number of QDs. \( \Pi_m \) denotes the sum of Coulomb interactions seen by a particle in dot \( l \) owing to other particles in dot \( j \) \((j \neq l)\), which can be occupied by zero, one, or two particles. \( p_m \) denotes the probability of such configurations. For a three-QD system \((l \neq j \neq f)\), there are nine \((3 \times 3)\) configurations, and the probability factors become \( p_1 = a_1a_f \), \( p_2 = b_1b_f \), \( p_3 = a_1b_f \), \( p_4 = c_1c_f \), \( p_5 = c_1b_f \), \( p_6 = b_1b_f \), \( p_7 = c_1b_f \), \( p_8 = c_1c_f \), where \( a_1 = 1 - (N_{\sigma,j} + N_{\sigma,-}) + c_j, \quad b_j = (N_{\sigma,j} + N_{\sigma,-}) \) is the intradot two particle correlation function, \( N_{\sigma,j} \) is the one particle occupation number. The interdot Coulomb interaction factors are \( \Pi_1 = 0, \Pi_2 = U_{1j}, \Pi_3 = U_{1f}, \Pi_4 = 2U_{1j}, \Pi_5 = 2U_{1f}, \Pi_6 = U_{1j} + U_{1f}, \Pi_7 = 2U_{1j} + U_{1f}, \Pi_8 = 2U_{1f} + U_{1j}, \) and \( \Pi_9 = 2U_{1j} + 2U_{1f} \). The sum of the probability factors \( p_m \) for all configurations is equal to 1, reflecting the fact that \( G_{\sigma,i}(\epsilon) \) satisfies the sum rule. The imaginary part of each resonant channel is \( \Gamma_i = (\Gamma_{iL} + \Gamma_{iR})/2 \) as a result of the coupling between the QDs and the electrodes in which the electron correlation effects are ignored owing to the consideration of the Coulomb blockade regime.

According to the retarded Green’s function of eq. (4), we need to know the occupation numbers \( N_{\sigma,i}(N_{\sigma,-}) \) and \( N_{\sigma} = c_i \); these can be obtained by solving the following equations self-consistently:

\[
N_{\sigma,i} = -\int \frac{d\epsilon}{\pi} \frac{\Gamma_{iL}f_L(\epsilon) + \Gamma_{iR}f_R(\epsilon)}{\Gamma_{iL} + \Gamma_{iR}} \text{Im}G_{\sigma,i}(\epsilon), \quad (5)
\]

\[
c_i = -\int \frac{d\epsilon}{\pi} \frac{\Gamma_{iL}f_L(\epsilon) + \Gamma_{iR}f_R(\epsilon)}{\Gamma_{iL} + \Gamma_{iR}} \text{Im}G_{i,j}(\epsilon). \quad (6)
\]

The values of \( N_{\sigma,i} \) and \( c_i \) are restricted between 0 and 1. The expression of two-particle retarded Green function of eq. (6) is

\[
G_{i,j}(\epsilon) = N_{\sigma,-} \sum_{m=1}^{N_{\sigma}} \frac{p_m}{\epsilon - E_j - \Pi_m + i\Gamma_j}.
\]

3. Results and Discussion

To study the direction-dependent heat current, we consider two sets of parameters (the cases of \( T_L > T_R \) and \( T_R > T_L \)): (a) \( \mu_L = E_F + e\Delta V, \mu_R = E_F, T_L = T_0 + \Delta T, \) and \( T_R = T_0 \), and (b) \( \mu_L = E_F, \mu_R = E_F - e\Delta V, T_L = T_0, \) and \( T_R = T_0 + \Delta T \). The average temperature of two side electrodes is \( (T_L + T_R)/2 = T_0 + \Delta T/2 \), which increases steadily with \( \Delta T \). Such a physical condition for average temperature was also employed to investigate the thermal rectification in a phonon junction system.\(^{25}\) A functional thermal rectifier requires a good thermal conductance for the case of \( T_L > T_R \), but a thermal insulator for the case of \( T_R > T_L \). On the basis of eqs. (2) and (3), a QD thermal rectifier requires not only highly asymmetrical coupling strengths between the QDs and the electrodes, but also strong electron Coulomb interactions between dots. To investigate the thermal rectification behavior, we have numerically solved eqs. (2) and (3) for three QD junctions for various system parameters.

The energy level of dot \( l \) is \( E_l = E_F + \alpha_l\Delta E \), where \(-1 < \alpha_l < 1\). \( \Delta E = 200^\circ \)C is used to reflect the energy level fluctuation of QDs. For simplicity, the intradot Coulomb interactions \( U_l = 600^\circ \)C are fixed, although the QD size fluctuation leads to the intradot Coulomb interaction variation. According to eq. (3), the direction-dependent heat currents are attributed to \( \Delta V \) and \( N_{\sigma,i} = N_{\sigma,-} = N_i \). The electrochemical potential \( \epsilon \Delta V \) is obtained by the Seebeck effect for the open circuit \( (J_0 = 0) \). Such an electrochemical potential is formed in response to the current generated by the temperature gradient. This electrochemical potential is known as the Seebeck voltage (Seebeck effect). The thermal power is defined as \( S = \Delta V/\Delta T \). Once \( \Delta V \) is solved, we then use eq. (3) to compute the heat current.

Figure 2 shows the direction-dependent heat current \( (Q) \) and rectification efficiency \( (\eta_0) \) as functions of \( \Delta T \) for various values of \( \Gamma_{iR} \), while keeping \( \Gamma_{iL} = \Gamma_{iC,LR} = \Gamma \). We have adopted the interdot Coulomb interactions \( U_{ij} = 300^\circ \)C and \( k_B T_0 = 25^\circ \)C. The energy levels of dots A, B, and C are chosen to be \( E_A = E_F - \Delta E/S, E_B = E_F + 2\Delta E/S, \) and \( E_C = E_F + 4\Delta E/S \), respectively. The heat currents have the
positive and negative signs corresponding, respectively, to the cases of $T_L > T_R$ and $T_R > T_L$. The former is the heat current from the left electrode to the right electrode $Q_{LR}$, and the latter is the heat current from the right electrode to the left electrode $Q_{RL}$. The rectification efficiency is defined as $\eta_{Q} = (Q_{LR}(\Delta T) - |Q_{RL}(\Delta T)|)/Q_{LR}(\Delta T)$. The rectification effect of heat currents shown in Fig. 2(a) can be clearly distinguished from that in Fig. 2(b). When dot A is fully blocked ($\Gamma_{AR} = 0$), the QD junctions exist a much higher $\eta_{Q}$, whereas the heat currents are seriously suppressed. Such a marked suppression implies that the heat currents through dots B and C are small because their resonant energy levels are far away from the Fermi energy level owing to large $U_{AC}$ and $U_{AB}$. The results shown in Fig. 2 indicate that it is crucial to well manipulate the coupling strengths between the dots and the electrodes in the optimization of thermal rectifiers.

In Fig. 2, we considered the homogenous interdot Coulomb interactions; however, it is difficult to have homogenous interdot Coulomb interactions in the multiple QD layout. The interdot Coulomb interaction fluctuation mainly arises from the size variation and the inhomogenous position distribution. We, therefore, examine the variation of $U_{AC}$ on heat currents. Figure 3 shows the heat current and rectification efficiency as functions of $\Delta T$ for various values of $U_{AC}$ at $\Gamma_{AR} = 0$ and $\Gamma_{AL} = 2\hbar$. Other parameters are the same as those in Fig. 2. The heat currents are suppressed by the strong interdot Coulomb interactions. This behavior is similar to the Coulomb blockade effect on the charge current.\(^{24}\) Nevertheless, a strong $U_{AC}$ leads to a higher rectification efficiency. Therefore, a more efficient thermal rectification effect can be observed in the high-density QD system. In such a case of strong intradot and interdot Coulomb interactions, the structure of charge current exhibits clear plateaus with respect to applied voltage. However, the structure of heat currents shown in Fig. 3 does not exhibit the plateaus with respect to temperature bias. This is because the electron distribution is broadened by temperature bias.\(^{24}\)

In Figs. 2 and 3 we considered the case of $E_C < E_F$. How $E_C$ affects the heat current and $\eta_{Q}$ is examined in Fig. 4 with $U_{AC} = 300\hbar$. Other parameters are the same as in Fig. 3.

When $E_C$ is tuned from $E_C = E_F + \Delta E/5$ to $E_C = E_F + 4\Delta E/5$, the heat current for $E_C = E_F + 2\Delta E/5$ is less than that for $E_C = E_F + 3\Delta E/5$. This result indicates that the magnitude of heat current is not adequately judged by only the separation between the energy levels of QDs and the Fermi energy level. When $E_C$ increases to $E_C = E_F + 4\Delta E/5$ from $E_C = E_F + 3\Delta E/5$, the heat currents and rectification efficiency are changed slightly. All of these complicated behaviors are attributed to the very nonlinear relationship between the heat currents and electrochemical potentials, which are strongly affected by the electron Coulomb interactions and the energy levels of each QD.

To further clarify the results in Fig. 4, the following expression of heat current, $Q = Q_{B} + Q_{C}$, is used:

$$Q_B = \frac{(1 - N_B) \cdot [(1 - 2N_A)(E_B - E_F) \cdot f_{LR}(E_B) + 2N_A(E_B + U_{AB} - E_F) \cdot f_{LR}(E_B + U_{AB})]}{\gamma_B \pi}, \tag{7}$$

$$Q_C = \frac{(1 - N_C) \cdot [(1 - 2N_A)(E_C - E_F) \cdot f_{LR}(E_C) + 2N_A(E_C + U_{AC} - E_F) \cdot f_{LR}(E_C + U_{AC})]}{\gamma_C \pi}, \tag{8}$$

\( \gamma_B, \gamma_C \) are the tunneling rates for the tunneling rate variation of dot A. The heat currents are distinguished from that in Fig. 2(b). When dot A is fully blocked ($\Gamma_{AR} = 0$), the QD junctions exist a much higher $\eta_{Q}$, whereas the heat currents are seriously suppressed. Such a marked suppression implies that the heat currents through dots B and C are small because their resonant energy levels are far away from the Fermi energy level owing to large $U_{AC}$ and $U_{AB}$. The results shown in Fig. 2 indicate that it is crucial to well manipulate the coupling strengths between the dots and the electrodes in the optimization of thermal rectifiers.
where $f_{LR}(e) = f_L(e) - f_R(e)$. Equations (7) and (8) are obtained by setting $c_1$ to zero owing to very large intradot Coulomb interactions and using a delta function to replace the Lorentzian function of resonant channels for $\Gamma \ll k_B T_0$ in eq. (3). In addition, only two resonant channels for each QD are considered. $N_A, N_B, N_C,$ and $\epsilon \Delta V$ are still solved by a full numerical solution [eqs. (2), (5), and (6)]. Figure 5 shows the heat current as a function of $\Delta T$ for $E_C = E_F + 3\Delta E/5.$ Other parameters are the same as those in Fig. 4. The dotted lines given by eqs. (7) and (8) match the full solutions very well. $Q_{LR}$ and $Q_{RL}$ are the nonlinear functions of $\Delta T$. Figure 5(b) shows the $Q_B$ and $Q_C$ calculated by using eqs. (7) and (8) for $Q_{LR}$ and $Q_{RL}$, respectively. On the basis of Fig. 5(b), the heat currents of Fig. 5(a) are mainly contributed by $Q_C$. For $Q_{LR}$, the behavior of heat current with respect to $\Delta T$ is affected by the factors of $N_A$ and $f_{LR}$. For $Q_{RL}$, the heat current behavior is affected only by the factor of $f_{LR}$ owing to the $\Delta T$-independent $N_A = 0.45$ for $\Gamma_{AR} = 0$. Because $f_{LR}$ is determined by the electrochemical potential $\epsilon \Delta V$, we will investigate this physical quantity, which is easily measured from the experimental point of view.

Figure 6 shows the electrochemical potential as a function of $\Delta T$ for different values of $k_B T_0$. Other parameters are the same as those in Fig. 5. The dot and dashed lines are the linear functions of $\Delta T$, whereas the solid lines are nonlinear with respect to $\Delta T$. Although the asymmetrical electrochemical potentials reveal some information about thermal rectification, it is difficult to judge the rectification efficiency from the measurement of electrochemical potentials in either the linear or nonlinear response case owing to the very nonlinear relationship between $Q$ and $\Delta V$. Therefore, the experimental results shown in ref. 17 are not sufficient evidence of the observation of the thermal rectification of a QD junction system. A large electrochemical potential yielded the temperature bias ($\Delta T$) is useful for generating electrical power. The results in Fig. 6 also imply that the charge current rectification with respect to temperature bias can be observed in the closed circuit ($I_e \neq 0$). An electrochemical potential is an essential quantity for charge carriers to deliver heat current; this is different from phonon carriers considered in the phonon junction system. To further illustrate the strong nonlinear relationship between the electrochemical potential and the heat current, we show the heat current and rectification efficiency in Fig. 7, where the curves correspond to those in Fig. 6. We observe that $n_0$ is larger at $k_B T_0 = 10\Gamma$ than at $k_B T_0 = 25\Gamma$. A comparison between Figs. 6 and 7 demonstrates the strong nonlinear relationship between $Q$ and $\Delta V$.

Finally, we calculate the thermal power as a function of $k_B T_0$ for different values of $\Delta T$ in Fig. 8. Other parameters are the same as those of Fig. 5. Thermal power plays a significant role in determining the figure of merit. However, much efforts has been devoted into the linear response regime of $\Delta T/T_0 \ll 1$. It is an expected result that the thermal powers are significantly affected by temperature bias for low $k_B T_0$. It is worth noting that the thermal power is enhanced with increasing temperature bias. Once $k_B T_0$ is high, the thermal powers become insensitive to the temperature bias. For some thermal devices, such as solid state coolers and electrical-power generators, their operation may be possible under the condition of $\Delta T/T_0 \geq 1$. Under such
Recently, the size and position of multiple germanium QDs has been precisely manipulated in SiO$_2$ amorphous insulators.$^{27}$ The experiment of ref. 27 indicates that it is possible to realize the system shown in Fig. 1 for examining the thermal rectification at high temperature. To avoid the phonon heat current, which seriously suppresses the heat rectification effect arising from electrons, the use of a scanning tunneling microscope (STM) in place of one of the electrodes is suggested since the vacuum layer between the STM and the amorphous insulator can block phonon heat current.

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4. Conclusions

In this theoretical study, we investigated the thermal rectification effects (TREs) of multiple QDs embedded into an amorphous insulator (phonon glass) with low heat conductivity. The rectification efficiency will be reduced in the presence of phonon heat current. TRE is significantly affected by the asymmetrical tunneling rates and strong interdot Coulomb interactions. Although the mechanism of thermal rectification is similar to that of charge current rectification, it is difficult to directly clarify the profound behavior of such a rectification effect owing to the very nonlinear relationship between the electrochemical potential and the heat current, which also leads to difficulty in judging the thermal rectification from thermal power measurements.

a situation, the thermal powers calculated in the linear response regime should be modified.

To enable practical applications, we must estimate the magnitudes of heat current and electrochemical potential to determine whether the effect of heat current is significant. We envision a thermal rectification device made of an array of multiple QDs (e.g., three-QD cells) with a 2D density $N_{\text{QD}} = 10^{11}$ cm$^{-2}$. For this device, the heat current density versus $k_B \Delta T = 16 \Gamma$ is given by Figs. 3 and 4 with $Q_0$ replaced by $N_{\text{QD}}Q_0$, which is approximately 482 W/m$^2$ if we assume $\Gamma = 0.5 \text{ meV}$. Under the temperature bias of $k_B \Delta T = 8 \text{ meV}$, the values of $\Delta V$ are, respectively, $-80$ and $40 \text{ meV}$ for $T_L > T_R$ and $T_R > T_L$, respectively, at temperature $k_B T_0 = 5 \text{ meV} = 60 K^0$ and intradot Coulomb energy $U = 300 \text{ meV}$.

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