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Study of ac hopping conductivity on one-dimensional nanometre systems

Xu Hui(徐 慧) and Song Yi-Pu(宋祎璞)

Department of Applied Physics, Central South University, Changsha 410083, China

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In this paper, we establish a one-dimensional random nanocrystalline chain model, we derive a new formula of ac electron–phonon–field conductance for electron tunnelling transfer in one-dimensional nanometre systems. By calculating the ac conductivity, the relationship between the electric field, temperature and conductivity is analysed, and the effect of crystalline grain size and distortion of interfacial atoms on the ac conductance is discussed. A characteristic of negative differential dependence of resistance and temperature in the low-temperature region for a nanometre system is found. The ac conductivity increases linearly with rising frequency of the electric field, and it tends to increase as the crystalline grain size increases and to decrease as the distorted degree of interfacial atoms increases.

Keywords: ac hopping conductivity, nanometre crystalline grain, distortion of interfacial atoms
PACC: 7210

1. Introduction

In 1984, Birringer and Gleiter[1] successfully synthesized a solid material with nanometre-sized grains. Nanometre particles are very tiny, and there are a great number of interfaces in the material. So the periodic arrangement of atoms in the system is greatly reduced. It is found experimentally that nanostructure materials exhibit various abnormal properties, such as high strength, high specific heat, high resistivity, high heat expansion coefficient, superplasticity, etc,[2−4] caused by interface effects and size effects. All of these exotic physical properties show that nanostructure materials hold the promise of extensive application. Now, research of these materials has become a new hotspot in the study of condensed-state physics.

The interfacial structure between the particles of nanometre materials packed bulk disorderly is very complicated, so it is very difficult to set up a mechanism of size effects and interface effects on the physicochemical properties. This may be the reason why experimental studies of nanostructure materials have been much more remarkably attractive than theoretical studies.

Conductance is an important performance of normal metals and alloys. The volume percentage of section in nanometre materials is very large, and destroys the lattice translation periodicity. The smaller the crystalline grain size is, the more serious the distortion of interfacial atoms is, and the shorter the average free path of atoms. Currently, research on nanometre conductance is in its early stages, and there are few experimental data and less theoretical computation. Ding *et al*[5,6] have calculated the conductivity of a one-dimensional (1D) nanocrystalline chain using the electron hopping conduction theory proposed by Miller and Abrahams.[7] They took into account the interaction between electrons and the electric field, but did not consider the scattering effect of atomic heat movement on electrons. Due to this reason and considering that a 1D problem is connected with clear physical ideas and effective mathematical methods, we propose a 1D nanocrystalline chain model, and develop a new method to compute eigenproblems of 1D nanocrystalline systems. Based on the mechanism of electron transfer, a new formula of electron conduction is derived, the conductivity of nanocrystalline system with 20000 atoms is calculated, and the effect of crystalline grain size and the distortion of interfacial atoms is analysed.

2.1D nanocrystalline chain model

In general, nanostructure materials are closely packed with ultrafine crystalline grains with sizes between 1–100nm. There are two kinds of atoms:[8] crystalline atoms which are arranged in periodic configurations, and interfacial atoms which are randomly packed. Furthermore, the relative directions of crys-

talline grains are also random. In a certain sense, the interfacial microstructure is the main factor that affects the properties of nanometre materials. We thus establish a 1D nanocrystalline chain model, as shown in Fig.1.

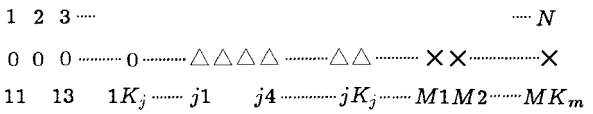


Fig.1. The 1D nanocrystalline chain model.

The symbols ◯, △ and × represent the crystalline atoms in a grain. This model contains M crystalline grains, and the sum index is indicated by j. jK_j represents that the jth crystalline grain has K_j atoms. If the total number of atoms is N, then

$$\sum_{i=1}^{M} K_i = N. \tag{1}$$

The Hamiltonian of this model can be written as

$$H = \sum_{n=1}^{M} \varepsilon_n \sum_{i=n1}^{nK_n} |i\rangle\langle i| + \sum_{l=1}^{N} \sum_{m=1(m\neq l)}^{N} t_{lm}|l\rangle\langle m|. \tag{2}$$

Here $|l\rangle$ is a group of known complete orthogonal basis functions. ε_j is a group of random numbers fluctuating irregularly between $-D/2$ and $D/2$, which represents the quality of each crystalline grain; namely, when electrons move in the crystalline grain, the potential field varies little, but when electrons stride over the grain boundary, it changes significantly. D is the distorted degree of the interfacial atoms. The larger D is, the more serious the distortion. t_{lm} denotes the action of neighbouring atoms. Concerning the nearest-neighbour(n-n) atoms and the next-n-n atoms, we take[9]

$$t_{lm} = -2/(|l-m|+1), \quad \text{when } |l-m| \leq H; \tag{3}$$

$$t_{lm} = 0, \quad \text{when } |l-m| \rangle H, \tag{4}$$

where H=2.

If λ is the eigenvalue of the Hamiltonian, and φ is its relevant eigenstate, then

$$H\varphi = \lambda\varphi. \tag{5}$$

Both the eigenvalue and eigenstate can be gained by solving the following matrix

$$\begin{bmatrix} \varepsilon_1-\lambda & -1 & -2/3 & & & & & & \\ -1 & \cdot & \cdot & \cdot & & 0 & & & \\ -2/3 & \cdot & \varepsilon_1-\lambda & -1 & -2/3 & & & & \\ & \cdot & -1 & \varepsilon_2-\lambda & -1 & -2/3 & & & \\ & & -2/3 & -1 & \cdot & \cdot & \cdot & & \\ & & & -2/3 & \cdot & \varepsilon_j-\lambda & -1 & -2/3 & \\ & & 0 & & \cdot & -1 & \cdot & -1 & \\ & & & & & -2/3 & -1 & \varepsilon_M-\lambda & \end{bmatrix} \begin{bmatrix} b_1 \\ b_2 \\ b_3 \\ \cdot \\ \cdot \\ \cdot \\ \cdot \\ b_N \end{bmatrix} = 0. \tag{6}$$

Using the negative eigenvalue theory of Dean and Martine[10] and the infinite order perturbation theory of Wu,[11] we have developed a new method to solve eigenvectors. This method is not only accurate but also efficient. It can compute the matrix of about 10^5 orders, and its computing error is below 10^{12}.

3. The ac conductance model for electron tunnelling in a nanometre system

3.1. The ac conductance model of electron tunnelling

In a nanometre system, the distortion of interfacial atoms and the crystalline grain size have a strong effect on the electronic structure. The electronic structure is different from that of a periodic lattice. The electron state expands in the whole energy band.[12] The electron wavefunction appears localized. With the increase of the distorted degree of interfacial atoms and the decrease of the crystalline grain size, the localization becomes strong. We will discuss the electronic structure of a nanometre system in another paper and do not discuss it in detail here.

Based on the interactions among phonons, electric field and electrons, the electron–phonon–field (EPF) conductance of electron tunnelling transfer is derived.

The ac electric field with electric intensity $E\cos\omega_{li}t$ is exerted on the nanometre system; here ω_{li} is the frequency of the electric field. When an electron absorbs the energies of phonons and photons, it hops from a low-energy to a high-energy localized state, and the difference between these energies is $\hbar\omega_{li}+W$, where W is the energy of phonons with $W=\hbar\omega_{ph}$, ω_{ph} being the frequency of phonons. The electron can also emit the energy of phonons and photons and hops from a high-energy to a lower-energy state. The electron hopping process is shown in Fig.2.

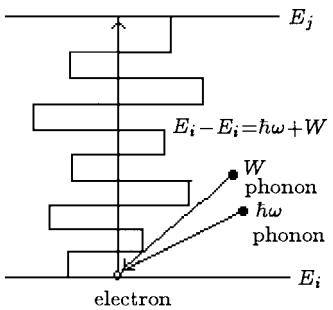


Fig.2. The EPF conductance of electron tunnelling transfer.

When absorbing the energies of photons and phonons, the electron hops between localized states. The hopping probability is determined by three factors:

(a) The tunnelling transfer probability of electrons. The electrons go through a series of potential barriers and wells, which fluctuate irregularly between $-D/2$ and $D/2$. The resonant transfer will take place when a certain condition is satisfied; at this moment, the tunnelling transfer probability is the greatest.

(b) The probability of finding a phonon whose energy is W. Since a phonon is a boson, its distribution is subordinated to the Bose–Einstein statistics. When the temperature is not too high, the probability is proportional to $e^{\frac{-W}{kT}}$, where k is the Boltzmann constant and T is temperature. It can clearly be seen that the probability decreases rapidly with the increasing energy of phonon.

(c) The vibration frequency of phonons, which assists electron hopping, namely the number of electron hoppings in unit time.

So electron hopping probability equals the product of three factors

$$P = U\mathrm{e}^{-W/kT}\nu_{Ph}\delta(E_i+\hbar\omega_{li}+W-E_j), \quad (7)$$

where ω_{li} is the frequency of ac electronic field, and U is the probability of electron tunnelling transfer.

3.2. The electron tunnelling transfer probability U

Using the transfer matrix method,[13,14] we calculate the hopping probability of electrons between different localized states. In the incident and emergence regions, the electron wavefunction can be obtained by solving the corresponding Schrödinger equation. The wavefunction is written respectively as follows

$$\phi_i(x) = \mathrm{e}^{ik_{ll}\rho}\begin{bmatrix} A_i\mathrm{e}^{ik_ix} \\ B_i\mathrm{e}^{ik_ix} \end{bmatrix}, \quad (8)$$

$$\phi_{i+n}(x) = \mathrm{e}^{ik_{ll}\rho}\begin{bmatrix} A_{i+n}\mathrm{e}^{ik_{i+n}x} \\ B_{i+n}\mathrm{e}^{ik_{i+n}x} \end{bmatrix}, \quad (9)$$

where k_{ll} represents the projection of the wave vector k on the plane perpendicular to the x-axis. In the potential barrier and well regions, we can solve the one-electron Schrödinger equations respectively and write the transfer matrix in terms of the condition that, at the interface of the potential barriers and potential wells, the wavefunction and its differential are continuous. Then we can obtain the transfer probability induced by the electrons when they transfer throughout the whole structure. The electron transfer process can be given by the equation

$$\begin{bmatrix} A_i\mathrm{e}^{ik_ix} \\ B_i\mathrm{e}^{ik_ix} \end{bmatrix} = M\begin{bmatrix} A_{i+n}\mathrm{e}^{ik_{i+n}x} \\ B_{i+n}\mathrm{e}^{ik_{i+n}x} \end{bmatrix}$$

$$= \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix}\begin{bmatrix} A_{i+n}\mathrm{e}^{ik_{i+n}x} \\ B_{i+n}\mathrm{e}^{ik_{i+n}x} \end{bmatrix}, \quad (10)$$

where M is the transfer matrix. In fact, in order to solve the above equation, we should take $B_{i+n}\mathrm{e}^{ik_{i+n}x}=0$. That is, there is only the transmission wave and no back wave in the emergence region. If we define the transmissivity as the ratio of transmission probability current density to incident probability current density, then U can be written as

$$U = \frac{k_{i+n}|A_{i+n}|^2}{k_i|A_i|^2} = \frac{k_{i+n}|M_{11}|^{-2}}{k_i}. \quad (11)$$

Considering the inversion time symmetry, the complex conjugate matrix of the eigenvector is also one of the solutions of the Schrödinger equation, i.e. $A_i^*\mathrm{e}^{-ik_ix}+B_i^*\mathrm{e}^{ik_ix}$ is the solution for the incident region, and $A_i^*\mathrm{e}^{-ik_{i+n}x}+B_i^*\mathrm{e}^{ik_{i+n}x}$ is the solution for the emergence region. Here B_i^* is the amplitude of the incident wave, A_i^* is the amplitude of the back wave and B_{i+n}^*

is the amplitude of the transmission wave. Then we have

$$\begin{bmatrix} A_i^* \mathrm{e}^{-ik_i x} \\ B_i^* \mathrm{e}^{ik_i x} \end{bmatrix} = \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix} \begin{bmatrix} A_{i+n}^* \mathrm{e}^{-ik_{i+n} x} \\ B_{i+n}^* \mathrm{e}^{ik_{i+n} x} \end{bmatrix}. \tag{12}$$

This equals

$$\begin{bmatrix} A_i \mathrm{e}^{ik_i x} \\ B_i \mathrm{e}^{ik_i x} \end{bmatrix} = \begin{bmatrix} M_{11}^* & M_{12}^* \\ M_{21}^* & M_{22}^* \end{bmatrix} \begin{bmatrix} A_{i+n}^* \mathrm{e}^{ik_{i+n} x} \\ B_{i+n}^* \mathrm{e}^{ik_{i+n} x} \end{bmatrix}, \tag{13}$$

where M must be satisfied with

$$M_{11}^* = M_{11},\ M_{12}^* = M_{12}. \tag{14}$$

Then it follows that

$$U = \frac{\det|M|}{|M_{11}|^2}. \tag{15}$$

4. The formula for ac conductivity in a nanometre system

Based on the EPF conductance model of electron tunnelling transfer we have derived, an electron interacts with photons and the electric field, and hops from site i to site j through a series of potential barriers and wells by absorbing or emitting a photon and/or a phonon. The electron hopping probability is

$$P = \frac{\det|M|}{|M_{11}|^2} \mathrm{e}^{-W/kT} \nu_{Ph} \delta(E_i + \hbar\omega_{li} + \hbar\omega_{Ph} - E_j). \tag{16}$$

By absorbing a photon, the electron hops from state $|E_i\rangle$ to state $|E_j\rangle$ and the exchanged energy in the process is

$$\begin{aligned} &\hbar\omega_{li} P f(E_i)[1 - f(E_i + \hbar\omega_{li} + \hbar\omega_{Ph})] \\ =&\hbar\omega_{li} \frac{\det|M|}{|M_{11}|^2} \mathrm{e}^{-W/kT} \nu_{Ph} f(E_i) \\ &\times [1 - f(E_i + \hbar\omega_{li} + \hbar\omega_{Ph})] \\ &\times \delta(E_i + \hbar\omega_{li} + \hbar\omega_{Ph} - E_j). \end{aligned} \tag{17}$$

By emitting a photon, the electron hops from state $|E_j\rangle$ to state $|E_i\rangle$ and the exchanged energy in this process is

$$\begin{aligned} &\hbar\omega_{li} P f(E_i + \hbar\omega_{li} + \hbar\omega_{Ph})[1 - f(E_i)] \\ =&\hbar\omega_{li} \frac{\det|M|}{|M_{11}|^2} \mathrm{e}^{-W/kT} \nu_{Ph} f(E_i + \hbar\omega_{li} + \hbar\omega_{Ph}) \\ &\times [1 - f(E_i)] \delta(E_i + \hbar\omega_{li} + \hbar\omega_{Ph} - E_j). \end{aligned} \tag{18}$$

By absorbing or emitting a photon, the exchanged energy is

$$\begin{aligned} &\hbar\omega_{li} P\{f(E_i)[1 - f(E_i + \hbar\omega_{li} + \hbar\omega_{Ph})] \\ &- f(E_i + \hbar\omega_{li} + \hbar\omega_{Ph})[1 - f(E_i)]\} \\ =&\hbar\omega_{li} \frac{\det|M|}{|M_{11}|^2} \mathrm{e}^{-W/kT} \nu_{Ph} \\ &\times [f(E_i) - f(E_i + \hbar\omega_{li} + \hbar\omega_{Ph})] \\ &\times \delta(E_i + \hbar\omega_{li} + \hbar\omega_{Ph} - E_j) \\ =&\hbar\omega_{li}(\hbar\omega_{li} + W) \frac{\det|M|}{|M_{11}|^2} \mathrm{e}^{-W/kT} \nu_{Ph} \left(- \frac{\partial f(E)}{\partial E} \right). \end{aligned} \tag{19}$$

The total energy exchanged is

$$\int \hbar\omega_{li}(\hbar\omega_{li} + W) \frac{\det|M|}{|M_{11}|^2} \mathrm{e}^{-W/kT} \nu_{Ph} \left(- \frac{\partial f(E)}{\partial E} \right) \mathrm{d}E, \tag{20}$$

where $f(E)$ is the Fermi function

$$f(E) = \frac{1}{\mathrm{e}^{(E - E_F)/kT} + 1}. \tag{21}$$

In terms of electrodynamics theory, the energy exchanged between the nanometre system and the ac electric field is equal to Joule energy:

$$\begin{aligned} \frac{1}{2} \sigma_{ac} E^2 = &\int \hbar\omega_{li}(\hbar\omega_{li} + W) \frac{\det|M|}{|M_{11}|^2} \\ &\times \mathrm{e}^{-W/kT} \nu_{Ph} \left(- \frac{\partial f(E)}{\partial E} \right) \mathrm{d}E. \end{aligned} \tag{22}$$

In the end, we obtain the ac conductivity of the nanometre system induced by the electron tunnelling transfer effect:

$$\begin{aligned} \sigma_{ac} = &\int \frac{2\hbar\omega_{li}(\hbar\omega_{li} + W)}{E^2} \frac{\det|M|}{|M_{11}|^2} \\ &\times \mathrm{e}^{-W/kT} \nu_{Ph} \left(- \frac{\partial f(E)}{\partial E} \right) \mathrm{d}E. \end{aligned} \tag{23}$$

It is easily seen that the electron hopping contributing to the conductance takes place near the Fermi surface. Therefore, the difference of energy between the original and final states, in which electron hops, is little, but the localized position of the electron differs significantly.[15] This means that the electron needs to hop far in order to find the original and final states whose energy is close to each other. At the limited temperature, the electron hops between the states near Fermi energy in the range of kT. Assuming that the electron hopping energies are equal to each other approximately, the ac conductive formula can be simplified as

$$\begin{aligned} \sigma_{ac} = &\frac{2\hbar\omega_{li}(\hbar\omega_{li} + W)}{E^2} \frac{\det|M|}{|M_{11}|^2} \\ &\times \mathrm{e}^{-W/kT} \nu_{Ph} \left(- \frac{\partial f(E)}{\partial E} \right) N(E_F) kT, \end{aligned} \tag{24}$$

where $N(E_F)$ is the electron state density near the Fermi energy.

5. Results and discussion

According to the formula of ac conductance, we calculate the conductivity of 1D nanocrystalline chain with 20 000 site points. Choosing the distortion parameter D and grain size parameter S as parameters, we analyse the effect of the distortion of interfacial atoms and crystal grain size on ac conductance of nanometre systems.

Figure 3 shows the relationship between the ac conductivity of nanometre systems and the frequency of the electric field. The systems corresponding to the curves a and b have identical temperature (T=100K) and identical distortion of interfacial atoms (D=1), but the crystal grain size S of a is different from that of b. a has 50–200 atoms in a grain whereas b has 800–1000 atoms. It can be seen that the conductivity of A and b increases linearly with rising electric field frequency, but their slopes are different, and the slope of b is larger than that of a.

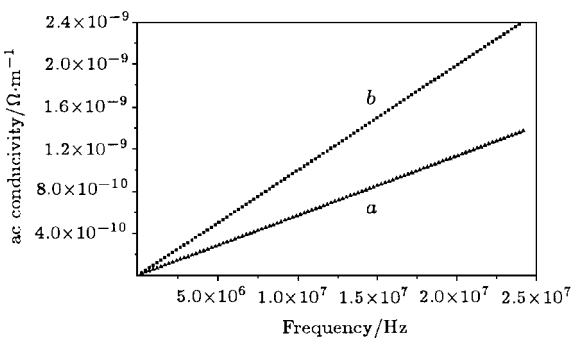


Fig.3. The relationship between ac conductivity and frequency of electric field: a, N=20 000, D=1, T=100K, S=50–200; b, N=20 000, D=1, T=100K, S=800–1000.

An experiment has been conducted, in which nanocrystalline NSi powder is pressed under a high pressure of 130MPa and its ac conductivity is measured for different frequencies of the electric field. The results show that $\sigma(\omega) = \omega^n$, and n varies from 0.65 at a low frequency to 1.6 at a high frequency.[16] The two indices n derived in this work according to the relationship between the ac conductivity and the frequency are 0.9696 and 0.9537, respectively, which is in good agreement with the experimental results. When the frequency of the electric field rises, the energy of the photon is increased. This reduces the phonon energy absorbed by the electron and means that the electron hopping probability increases. Therefore, the ac conductivity of nanometre systems increases as the frequency of the electric field rises.

Figure 4 demonstrates the relationship between ac conductivity and temperature, where three curves are drawn for different nanocrystalline systems. The nanometre chains a, b and c have the same number of atoms, but the crystalline grain size and the distortion of interfacial atoms are different from each other. From this figure, it can be seen that they have a similar relationship of conductivity and temperature. In the low-temperature region, the nanometre system presents the characteristic of negative differential dependence of resistance and temperature, i.e. the conductivity increases with rising temperature. In contrast, the conductivity decreases with rising temperature in the high-temperature region. This fits well with the experimental conclusion of Savvides *et al*[17] about the nanometre material of Cu–SiO_2 and that of Liu *et al*.[18] Compared with the result obtained by Ding[5] who used the $M - A$ formula, our result is more consistent with the experimental data. This phenomenon is connected with the electron hopping between localized states and can be explained as follows. In the low-temperature region, the heat movement of atoms is not violent and this movement will be stimulated gradually when the temperature is increased. This will be beneficial for electron hopping and makes the conductivity rise. But in the high-temperature region, the heat movement of atoms is so violent that it will cause scattering action on electron hopping, reducing the conductivity.

The temperature corresponding to the maximum value of conductivity is different for different systems. It is determined by the properties of the system itself, such as the size of crystalline grains and the distortion of interfacial atoms, etc.

In addition, it can be seen from Fig.4 that the distortion of interfacial atoms and the size of crystalline grains have great effect on ac conductance of nanometre systems. Comparing nanometre chains a and c, we see that the distorted parameter D is different, but the grain size S is the same, and the conductivity of a is obviously larger than that of b. This shows that the ac conductivity of a nanometre system tends to decrease as the distorted degree of interfacial atoms is increased. This is caused by the fact that the heat movement of lattice atoms has a scattering action on the directional movement of electrons. The more serious the distortion of interfacial atoms is, the more obvious the scattering action.

In Fig.4, comparing nanometre chains b and c, we see that parameter D in the two chains is identical, but S is different, and the crystalline grain size of chain c is much larger than that of chain b. The conductivity of c is larger than that of b. This shows that ac conductivity of nanometre systems tends to rise as crystalline grain size is increased. This result is similar to the experimental conclusion of Gleiter.[8] The reason is that the smaller the crystalline grain size is, the larger the percentage of interfacial atoms, the more serious the destroyed degree of periodical potential field, and the more remarkable the scattering action of distorted crystalline atoms on electrons. In a certain case, the crystalline grain size has a stronger effect than the distortion of crystalline interfacial atoms on ac conductance of nanometre systems.

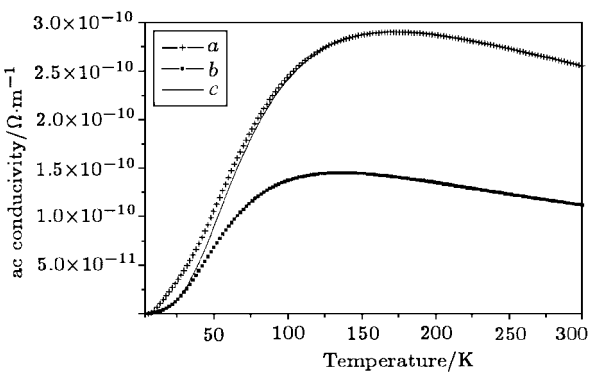


Fig.4. The relationship between the ac conductivity and the temperature: a, N=20 000, D=0.5, S=50–200; b, N=20 000, D=1, S=50–200; c, N=20 000, D=1, S=800–1000.

6. Conclusion

From the above discussion, we can draw conclusions as follows:

(1) The ac conductive characteristic of a nanometre system is completely different from that of a crystalline system with large crystals. The electron hopping contribution to the conductance takes place near the Fermi surface. The difference of electron hopping energies between the original and final states is small, but the distance of the positions of states may be great.

(2) The ac conductivity of a nanometre system increases linearly with the increase of electric field frequency. The slopes of conductivity curves are different for different systems.

(3) In the low-temperature region, a nanometre system presents the characteristic of negative differential dependence of resistance on temperature, i.e. the conductivity increases with rising temperature. In contrast, its conductivity decreases with rising temperature in the high-temperature region. The temperature corresponding to the maximum value of conductivity is different for different systems.

(4) The size of crystalline grains and the distortion of interfacial atoms have a great effect on the ac conductance of nanometre systems. The conductivity tends to rise as the crystalline grain size is increased, and to decrease as the distorted degree of interfacial atoms is increased.

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