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Jump frequency of silver ions for diffusion in α-Ag_2Te

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The diffusion process of silver ions in α-Ag_2Te is investigated. It is assumed that Ag ions occupy the tetrahedral sites and octahedral sites with different probability. To give an explanation of the Havens ratio obtained by a molecular-dynamics calculation, a theory of caterpillar mechanism proposed by Yokota [J. Phys. Soc. Jpn. **21**, 420 (1966)] is used. It is shown that the ratio of the frequency of a single jump to that of cooperative jumps increases with increasing temperature.

I. INTRODUCTION

Okazaki[1,2] has measured the self-diffusion coefficient D and the mobility μ of silver ions in silver chalcogenides using a tracer technique. The mobility is related to the diffusion coefficient by the Einstein relation $\mu = eD/kT$. His experimental results have shown that the Einstein relation does not hold in silver chalcogenides. We can express the deviation from the Einstein relation quantitatively by introducing a correlation factor f as follows:

$$f = eD/\mu kT . \tag{1}$$

This correlation factor is called the Havens ratio. The f value of Ag_2Te is fairly large in comparison with those of other silver chalcogenides, Ag_2S and Ag_2Se. This has been interpreted in the following expression.[2] The value of the self-diffusion coefficient of α-Ag_2Te is almost the same as that of α-Ag_2S and α-Ag_2Se, while the value of the ionic conductivity of α-Ag_2Te is much smaller than that of α-Ag_2S and α-Ag_2Se. These facts lead to the differences in the f value.

In nearly perfect crystals, atomic diffusion is always connected with the existence of lattice defects. The most common types of diffusion in solids are the vacancy and interstitial mechanisms.[3-5] However, the crystal structure of superionic conductors is far from being perfect. In superionic conductors, the mobile ions are distributed randomly over the available sites in the crystal lattice. Then, it is not appropriate to rely upon the existing diffusion theory to explain the deviation from the Einstein relation in superionic conductors. Yokota[6] has proposed a caterpillar theory for the diffusion of silver ions to give an account of the remarkable deviation from the Einstein relation in silver chalcogenides. He has supposed that an ion on a site is able to jump not only into a vacant neighboring site but also into an occupied one, inducing the ion on the latter site to make a jump. Okazaki[2] has applied the caterpillar theory to interpret the observed f value of α-Ag_2Te, assuming that silver ions occupy the tetrahedral and octahedral sites with equal probability.

Recently we have investigated the structural and dynamical properties of α-Ag_2Te using a molecular-dynamics (MD) simulation.[7] The MD calculations have suggested that the Ag ion stays at the tetrahedral site most of the time and moves to its neighboring tetrahedral site through the vicinity of the octahedral site. This is in good agreement with the x-ray experiment.[8] Then, it is not appropriate to assume that silver ions occupy the tetrahedral and octahedral sites with equal probability.

In this paper, we study the diffusion process of silver ions in α-Ag_2Te. We apply the Yokota caterpillar-mechanism theory to interpret the Havens ratio obtained by the MD calculation. We assume that two kinds of sites—tetrahedral and octahedral—are available for silver ions. We also assume that silver ions occupy these two sites with a different probability, considering the results of the MD calculation and the x-ray experiment. In Sec. II we calculate the self-diffusion coefficient of Ag ions by investigating their spontaneous and induced jumps. In Sec. III we give the calculation of the current associated with those jumps. In Sec. IV we discuss the cooperative motion of Ag ions in α-Ag_2Te, comparing the calculation by the caterpillar-mechanism theory with the results by the MD calculation.

II. SELF-DIFFUSION COEFFICIENT

As stated in the Introduction, MD calculations have suggested that Ag ions in Ag_2Te do not jump from tetrahedral site to tetrahedral site directly on the $(\frac{1}{4}00)$ plane, but jump from tetrahedral site to tetrahedral site through the vicinity of the octahedral site along zigzag paths. These tetrahedral and octahedral sites are shown in Fig. 1. It is not easy to apply the caterpillar mechanism to these zigzag paths. For simplicity we consider a

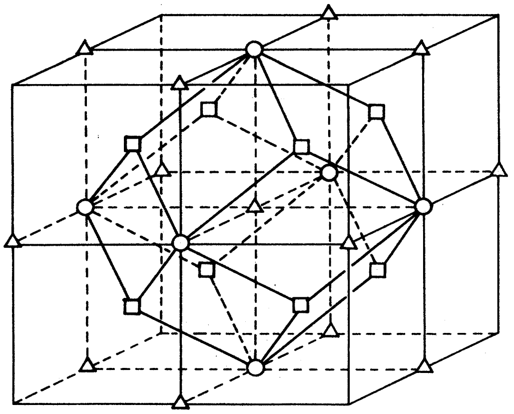

FIG. 1. Configuration of Ag_2Te. Squares and circles show the tetrahedral and octahedral sites, respectively. Triangles show the Te ions which construct the fcc lattice.

system which has at equal intervals alternating A and B sites which correspond to a tetrahedral and an octahedral site, respectively, on the straight line. We denote the jump frequency from a B site to a vacant neighboring site A by $\nu_{BA}(1)$ and that to an occupied site A by $\nu_{BA}(2)$. Similarly, we denote the jump frequency from an A site to a vacant neighboring site B by $\nu_{AB}(1)$ and that to an occupied site B by $\nu_{AB}(2)$. It is required that silver ions occupy site A with probability P_A and site B with probability P_B. We consider the cooperative jumps shown in Fig. 2. If a tracer ion T is put on the zeroth site and makes a spontaneous jump to the neighboring first site which is occupied by a Ag ion, the Ag ion on the first site will be induced to make a jump to the second site. If all the sites from 0 to $n-1$ are occupied and the nth site is vacant (case of Fig. 2), the successive jumps will continue until a Ag ion on the $(n-1)$th site makes a jump to the nth site. Such n successive jumps will contribute n steps to the electrical conduction, while contributing only one step to the self-diffusion of a tracer ion T.

When the zeroth site is a site A, the probability per unit time that the tracer ion on the zeroth site makes a spontaneous jump 0→1, the jump frequency $\nu_{sp}(A)$, is equal to

$$\nu_{sp}(A)=\nu_{AB}(1)(1-P_B)+\nu_{AB}(2)P_B \ . \tag{2}$$

Similarly, when the zeroth site is a site B, the jump frequency $\nu_{sp}(B)$ of the tracer ion on the zeroth site is equal to

$$\nu_{sp}(B)=\nu_{BA}(1)(1-P_A)+\nu_{BA}(2)P_A \ . \tag{3}$$

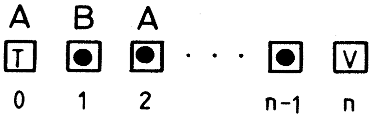

FIG. 2. Cooperative jumps associated with the spontaneous jump 0→1 of tracer ion T. The nth site is vacant. This is the case in which the zeroth site is on a site A.

Then, a spontaneous jump frequency ν_{sp} is given by

$$\nu_{sp}=c_A\nu_{sp}(A)+c_B\nu_{sp}(B) \ , \tag{4}$$

where c_A and c_B are the ratios of concentration of site A and site B, respectively, and $c_A+c_B=1$. Here we take $c_A=c_B=0.5$ in our model.

Now we calculate the jump frequency per unit time that the tracer ion on the zeroth site makes an induced jump 0→1. We consider the cooperative jumps shown in Fig. 3. If all of the sites from $-m$ to -1 are occupied by Ag ions, a spontaneous jump of Ag ion on the $-m$th site to the $-(m-1)$th site will induce successive jumps and lead a jump 0→1 of the tracer ion on the zeroth site. We have to calculate the two cases where the zeroth site is on an A site and the zeroth site is on a B site.

We first consider the case where the zeroth site is on an A site. The probability that all the sites from $-m$ to -1 are occupied is $P_B^{(m+1)/2}P_A^{(m-1)/2}$ for odd m. When we set $m=2l+1$ $(l=0,1,2,\ldots)$, the probability per unit time that the tracer ion T makes an induced jump 0→1 is

$$\sum_{l=0}^{\infty}\nu_{BA}(2)P_B^{l+1}P_A^{l}=\frac{P_B}{1-P_AP_B}\nu_{BA}(2) \ .$$

For even m, it is obtained as

$$\sum_{l=1}^{\infty}\nu_{AB}(2)P_B^{l}P_A^{l}=\frac{P_AP_B}{1-P_AP_B}\nu_{AB}(2) \ .$$

Then, the probability per unit time that the tracer ion T makes an induced jump 0→1 for the case where the zeroth site is on an A site is given by

$$\nu_{ind}(A)=\frac{P_B}{1-P_AP_B}[\nu_{BA}(2)+\nu_{AB}(2)P_A] \ .$$

Similarly, the probability per unit time that the tracer ion T makes an induced jump 0→1 for the case where the zeroth site is on a B site is given by

$$\nu_{ind}(B)=\frac{P_A}{1-P_AP_B}[\nu_{AB}(2)+\nu_{BA}(2)P_B] \ .$$

When we sum up these two cases, we have the induced jump frequency ν_{ind} as follows:

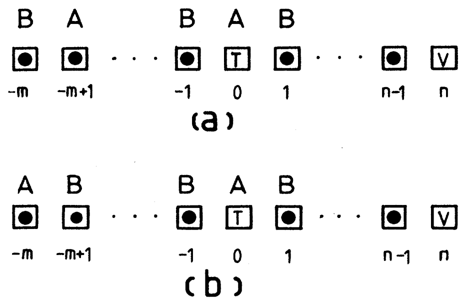

FIG. 3. A diagram which contributes to the induced jump frequency ν_{ind}. All of the sites from $-m$ to -1 are occupied. The figure shows the case where the tracer ion T is on the A site, in (a) for odd m and (b) for even n.

$$\nu_{\mathrm{ind}}=c_A\nu_{\mathrm{ind}}(A)+c_B\nu_{\mathrm{ind}}(B)=\frac{P_AP_B[\nu_{AB}(2)+\nu_{BA}(2)]+\nu_{BA}(2)P_B+\nu_{AB}(2)P_A}{2(1-P_AP_B)}\ . \tag{5}$$

While the jump frequency of a Ag ion is proportional to exp(−activation energy/kT), the occupation probability is proportional to exp(−on-site energy/kT). Then we have the following relation:

$$\nu_{AB}(1)/\nu_{BA}(1)=P_B/P_A\ . \tag{6}$$

The total jump frequency ν of a tracer ion is given by the sum of ν_{sp} and ν_{ind} as follows:

$$\nu=\frac{\nu_{1AB}}{2}\left\{1-P_B+\frac{P_B}{\alpha}+\frac{P_A}{P_B}(1-P_A)+\frac{P_A^2}{\beta P_B}+\frac{1}{1-P_AP_B}\left[P_AP_B\left[\frac{1}{\alpha}+\frac{P_A}{\beta P_B}\right]+\frac{P_A}{\beta}+\frac{P_A}{\alpha}\right]\right\}, \tag{7}$$

with

$$\alpha=\nu_{AB}(1)/\nu_{AB}(2),\quad \beta=\nu_{BA}(1)/\nu_{BA}(2)\ .$$

The self-diffusion coefficient D of the tracer ion can be expressed in the term of the jump frequency ν as

$$D=\nu a^2\ , \tag{8}$$

where a is the distance between two neighboring sites.

III. CURRENT

Next we have to calculate the current. The calculation is more complicated than that of the jump frequency. The total current J is made up of the following two parts: one is from the spontaneous jump $0\rightarrow1$ of the tracer ion and the other is from the induced jump $0\rightarrow1$ of the tracer ion on the zeroth site.

First we calculate the charge transport per unit time associated with the spontaneous jump $0\rightarrow1$ of the tracer ion T. We consider the case where the zeroth site is on an A site. When the first site is vacant, the contribution to the current associated with the spontaneous jump $J_{\mathrm{sp}}(A)$ is

$$ea\nu_{AB}(1)(1-P_B)\ .$$

When the first to $(n-1)$th sites are occupied but the nth site is vacant (see Fig. 2), the contribution to $J_{\mathrm{sp}}(A)$ is

$$ea\nu_{AB}(2)\sum_{l=1}^{\infty}(2l+1)P_A^lP_B^l(1-P_B)=ea\nu_{AB}(2)(1-P_B)\left[\frac{2P_AP_B}{(1-P_AP_B)^2}+\frac{P_AP_B}{1-P_AP_B}\right]$$

for odd $n=2l+1$ and

$$ea\nu_{AB}(2)\sum_{l=1}^{\infty}2lP_A^{l-1}P_B^l(1-P_A)=ea\nu_{AB}(2)2(1-P_A)P_B/(1-P_AP_B)^2$$

for even $n=2l$. Then the total current $J_{\mathrm{sp}}(A)$ by the spontaneous jump $0\rightarrow1$ of the tracer ion for the case where the zeroth site is on an A site is given by

$$J_{\mathrm{sp}}(A)=ea\nu_{AB}(1)(1-P_B)+\frac{ea\nu_{AB}(2)}{1-P_AP_B}(2-P_AP_B+P_A)P_B\ .$$

Similarly, the current $J_{\mathrm{sp}}(B)$ for the case where the zeroth site is on a B site is obtained as

$$J_{\mathrm{sp}}(B)=ea\nu_{BA}(1)(1-P_A)+\frac{ea\nu_{BA}(2)}{1-P_AP_B}(2-P_AP_B+P_B)P_A\ .$$

Then, the current J_{sp} accompanied by the spontaneous jump of the tracer ion is given by

$$\begin{aligned}J_{\mathrm{sp}}&=C_AJ_{\mathrm{sp}}(A)+C_BJ_{\mathrm{sp}}(B)\\&=\frac{ea}{2}\left\{\nu_{AB}(1)(1-P_B)+\nu_{BA}(1)(1-P_A)+\frac{1}{1-P_AP_B}[\nu_{AB}(2)(2-P_AP_B+P_A)P_B+\nu_{BA}(2)(2-P_AP_B+P_B)P_A]\right\}\ .\end{aligned} \tag{9}$$

Next we calculate the current J_{ind} associated with the induced jump $0\rightarrow1$. We consider the case shown in Fig. 4, where the $-m$th to the $(n-1)$th sites are occupied, but the nth site is vacant. We have to calculate the contribution from the $m+n$ successive jumps starting from the spontaneous jump $(-m)$th site$\rightarrow(-m+1)$th site and ending at the nth site. We assume the case where the zeroth site is on an A site. According to the odd or even numbers of m and n, we investigate the following four cases.

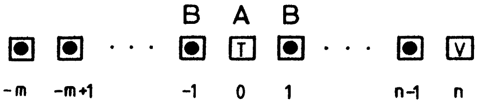

FIG. 4. A diagram which contributes to the current J_{ind} associated with the induced jump 0⟶1. The $(-m)$th to the $(n-1)$th sites are occupied but the nth site is vacant.

(1) For m even and n even, the probability that all the sites from the $(-m)$th site to the $(n-1)$th site are occupied, but the nth site is vacant, is

$$(P_A)^{(m+n-2)/2}(P_B)^{(m+n)/2}(1-P_A)\ .$$

Taking $m=2l$ $(l=1,2,\ldots)$ and $n=2k$ $(k=1,2,\ldots)$ we have the following contribution to J_{ind}:

$$ae\nu_{AB}(2)(1-P_A)\sum_{k=1}^{\infty}\sum_{m=1}^{\infty}2(k+l)P_A^{k+l-1}P_B^{k+l}$$
$$=4ae\nu_{AB}(2)P_AP_B^2(1-P_A)/(1-P_AP_B)^3\ .$$

(2) For m odd and n even, taking $m=2k-1$ $(k=1,2,\ldots)$ and $n=2l$ $(l=1,2,\ldots)$, the contribution to J_{ind} is given by

$$ae\nu_{BA}(2)\sum_{k=1}^{\infty}\sum_{l=1}^{\infty}(2k+2l-1)P_A^{k+l-2}P_B^{k+l}(1-P_A)$$
$$=ae\nu_{BA}(2)\left[\frac{4(1-P_A)P_B^2}{(1-P_AP_B)^3}-\frac{(1-P_A)P_B^2}{(1-P_AP_B)^2}\right]\ .$$

(3) For m even and n odd, taking $n=2l-1$ $(l=1,2,\ldots)$ and $m=2k$ $(k=1,2,\ldots)$, the contribution to J_{ind} is given by

$$ae\nu_{AB}(2)\sum_{k=1}^{\infty}\sum_{l=1}^{\infty}(2k+2l-1)P_A^{(2k+2l-2)/2}P_B^{(2k+2l-2)/2}(1-P_B)=ae\nu_{AB}(2)\left[\frac{4(1-P_B)P_AP_B}{(1-P_AP_B)^3}-\frac{(1-P_B)P_AP_B}{(1-P_AP_B)^2}\right]\ .$$

(4) For m odd and n odd, taking $n=2l-1$ $(l=1,2,\ldots)$ and $n=2k-1$ $(k=1,2,\ldots)$, the contribution to J_{ind} is given by

$$ae\nu_{BA}(2)\sum_{k=1}^{\infty}\sum_{l=1}^{\infty}2(k+l-1)P_A^{k+l-2}P_B^{k+l-1}(1-P_B)=2ae\nu_{BA}(2)P_B(1-P_B)(1+P_AP_B)/(1-P_AP_B)^3\ .$$

Summing up these four contributions, we obtain $J_{\text{ind}}(A)$ for the case where the zeroth site is on an A site,

$$J_{\text{ind}}(A)=\frac{ae}{(1-P_AP_B)^3}[4\nu_{AB}(2)P_AP_B^2(1-P_A)+\nu_{BA}(2)(3+P_AP_B)(1-P_A)P_B^2$$
$$+\nu_{AB}(2)(1-P_B)P_AP_B(3+P_AP_B)+2\nu_{BA}(2)(1-P_B)P_B(1+P_AP_B)]\ .$$

Similarly, the current $J_{\text{ind}}(B)$ for the case where the zeroth site is on a B site is obtained by exchanging the subscripts A and B in the above equation. Then the current J_{ind} associated with the induced jump 0⟶1 is obtained as

$$J_{\text{ind}}=C_AJ_{\text{ind}}(A)+C_BJ_{\text{ind}}(B)=\frac{ae}{2(1-P_AP_B)^3}\{4P_AP_B[\nu_{AB}(2)P_B(1-P_A)+\nu_{BA}(2)P_A(1-P_B)]$$
$$+(3+P_AP_B)[\nu_{BA}(2)(1-P_A)P_B^2+\nu_{AB}(2)(1-P_B)P_A^2]$$
$$+(3+P_AP_B)P_AP_B[\nu_{AB}(2)(1-P_B)+\nu_{BA}(2)(1-P_A)]$$
$$+2(1+P_AP_B)[\nu_{BA}(2)(1-P_B)P_B+\nu_{AB}(2)(1-P_A)P_A]\}\ . \tag{10}$$

Finally the total current J associated with the jump 0⟶1 of tracer ion T is obtained by summing up J_{sp} and J_{ind} as follows:

$$J=\frac{ae\nu_{AB}(1)}{2}\left[1-P_B+\frac{P_A}{P_B}(1-P_A)+\frac{1}{1-P_AP_B}\left[\frac{1}{\alpha}(2-P_AP_B+P_A)P_B+\frac{P_A^2}{\beta P_B}(2-P_AP_B+P_B)\right]\right.$$
$$+\frac{4P_A}{(1-P_AP_B)^3}\left[\frac{P_B^2}{\alpha}(1-P_A)+\frac{P_A^2}{\beta}(1-P_B)\right]+\frac{P_A(P_A+P_B)}{(1-P_AP_B)^2}\left[\frac{4}{1-P_AP_B}-1\right]\left[\frac{1-P_B}{\alpha}+\frac{1-P_A}{\beta}\right]$$
$$\left.+\frac{2P_A}{(1-P_AP_B)^2}\left[\frac{2}{1-P_AP_B}-1\right]\left[\frac{1-P_A}{\alpha}+\frac{1-P_B}{\beta}\right]\right]\ . \tag{11}$$

Here we have used Eq. (6) to express Eqs. (7) and (11).

IV. HAVENS RATIO AND DISCUSSIONS

The residence time of silver ions at the tetrahedral and octahedral sites have been calculated by a molecular-dynamic (MD) method.[7] The residence time can be expressed by the time steps taken for its residence. The time steps which have been calculated by MD are shown in Table I. We can define the ratio of residence time steps as follows:

$$z_T=y_T/(y_T+y_0),\quad z_0=y_0/(y_T+y_0)\ . \tag{12}$$

It is clear that

$$z_T+z_0=1\ . \tag{13}$$

There are eight Ag ions and eight tetrahedral sites in a unit cell. Therefore, we can take the occupation probability P_A to be equal to z_T. When we take into account that there are four octahedral sites in a unit cell, we have

$$8P_A+4P_B=8\ . \tag{14}$$

Then we have

$$P_B=2z_0\ . \tag{15}$$

This is reasonable to consider that the number of Ag ions in a unit cell is twice the number of octahedral sites. Kobayashi *et al.*[7] have calculated the Havens ratio by a MD method and have had 0.5 as its value. Their result that the Havens ratio of Ag ions in α-Ag_2Te has not been dependent upon the temperature has agreed well with the experimental result.[2] The Havens ratio is defined by

$$f_H=eav/J\ . \tag{16}$$

By making use of Eqs. (7) and (11)–(15), the values of parameters α and β are estimated so that we may obtain the value of Havens ratio by MD calculations, $f_H=0.5$. In Fig. 5 the α-β map for $f_H=0.5$ is shown for two temperatures. $\nu_{AB}(1)$ and $\nu_{AB}(2)$ are the jump frequencies from a site A to a vacant and an occupied neighboring site B, respectively. Then the former will exhibit a characteristic resembling single hopping and the latter a characteristic resembling a cooperative jump . To realize $f_H=0.5$, some cooperative jumps are required. For the case of a small value of $\nu_{AB}(2)$, a large value of $\nu_{BA}(2)$ has to guarantee a cooperative jump and vice versa. Figure 5 shows that the smaller of the two parameters α and β is dominant to get the value of $f_H=0.5$ for the case of a small value of $\nu_{AB}(2)$ or $\nu_{BA}(2)$.

TABLE I. The temperature dependence of the Ag-ion density in α-Ag_2Te by MD calculations (Ref. 7). y_T and y_0 are the rates of time steps of how long Ag ions stay at tetrahedral and octahedral sites, respectively. The rate of y_0 is taken as unity.

Temperature (K)	y_T	y_0
550	64.75	1
650	18.45	1
750	7.17	1
850	5.29	1

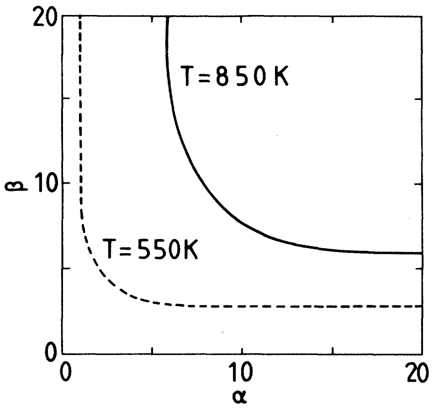

FIG. 5. α-β map of $f_H=0.5$ for two temperatures, 550 and 850 K.

Here we consider the value of $\nu_{AB}(2)/\nu_{AB}(2)$. An energy-barrier diagram where sites A and B are occupied is illustrated schematically in Fig. 6. Δ' and δ' are the barrier height. The barrier height will be higher than that of the case where the neighboring site is empty. Therefore the jump frequencies $\nu_{AB}(2)$ and $\nu_{BA}(2)$ will get smaller than those of the latter case. $\nu_{AB}(2)$ is proportional to $\exp(-\beta\Delta')$ and $\nu_{BA}(2)$ is proportional to $\exp(-\beta\delta')$. Then we have

$$\nu_{AB}(2)/\nu_{BA}(2)=\exp[-\beta(\Delta'-\delta')]\ . \tag{17}$$

On the other hand, the occupation probability is proportional to exp(on-site energy). Namely P'_A is proportional to $\exp(-\beta E'_A)$ and P'_B is proportional to $\exp(-\beta E'_B)$. E'_A and E'_B are the energies of sites A and B, respectively. Then we have

$$P'_B/P'_A=\exp[-\beta(E'_B-E'_A)]\ . \tag{18}$$

From Eqs. (17) and (18), we have

$$\nu_{BA}(2)/\nu_{AB}(2)=P'_A/P'_B\ . \tag{19}$$

Using Eqs. (6) and (19), we have

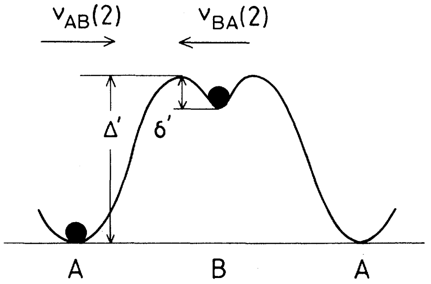

FIG. 6. An energy-barrier diagram where sites A and B are occupied. Δ' and δ' are the barrier heights.

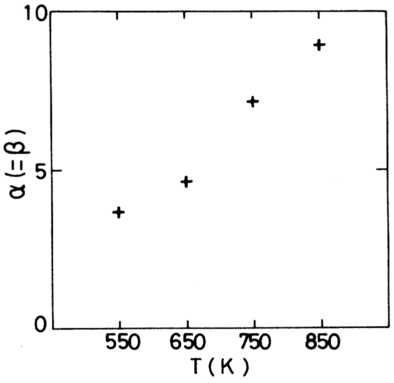

FIG. 7. The temperature dependence of α ($=\beta$).

$$\alpha/\beta=[\nu_{AB}(1)/\nu_{AB}(2)]/[\nu_{BA}(1)/\nu_{BA}(2)]$$
$$=P_B P'_A/P_A P'_B \ . \tag{20}$$

If an energy of each site is independent of the configuration diagram where the neighboring site is occupied or not, we can replace P'_A/P'_B with P_A/P_B and then we will have the equality $\alpha=\beta$. Since the Ag-density distribution of tetrahedral and octahedral sites in α-Ag$_2$Te is given by the statistical average as shown in Table I and does not depend on the instantaneous configuration diagram, the assumption $\alpha=\beta$ will be right. Then we assume the equality $\alpha=\beta$. Figure 7 shows the temperature dependence of α for the case where the equality of $\alpha=\beta$ is assumed. The value of α increases with increasing temperature. The potential barrier will decrease relatively in comparison with the kinetic energy of a mobile ion with the temperature increase and then the jumping process of Ag ions will be frequent. When temperature increases, the randomness of a system enlarges and then the frequency of a single jump increases relatively, compared with a cooperative jump. This shows the temperature dependence of α in Fig. 7.

To summarize, we have investigated the cooperative motion of Ag ions in α-Ag$_2$Te using Yokota's caterpillar-mechanism theory. The theory has been applied with some modification to the system so that it may be appropriate to the results of MD calculation. The temperature dependence of the jump frequency has been studied by making use of the results of the MD calculation.

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