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Positron Lifetime Calculation for Defects in Graphite

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Positron lifetime calculation has been performed for a vacancy and vacancy clusters in graphite and the comparison with experimental result was also made. Defect structure was obtained in a model graphite lattice after the relaxation of whole lattice using the molecular dynamics method, where the atomic potential presented by Andribet et al. was used. For the defect structure thus obtained positron lifetime was calculated under the atomic superposition method. Positron lifetimes 204 psec and 222 psec were obtained for the graphite matrix and a single vacancy, respectively, which can be compared with the experimental results 208psec and 233psec. For larger vacancy clusters, namely, vacancy loops lifetime calculation was also made and the increasing tendency with the number of vacancies in a cluster appeared. This is consistent with the experimental result in the higher annealing temperature region, where the increase of positron lifetime is seen, pobably corresponding to the clustering of mobile vacancies.

1. Introduction

Graphite has recently become one of the important candidates for the plasma facing materials in a fusion reactor, because of its high performance at high temperature region, such as high thermal conductivity and low Z character[1]. Hydrogen recycling in graphite is a very important problem for the operation of the fusion reactor and is tightly connected to the defect behaviors in surface region of graphite, and then defect properties in graphite must be investigated more in detail[2)-10)]. On the other hand, positron annihilation technique is very sensitive to vacancy type defects and positron lifetime calculation has recently become possible, which enables the comparison between the experiment and the calculation[11)12)]. In the present study positron lifetime at defects in graphite will be calculated and will be compared with experimental results.

2. Method of Calculation

Model graphite lattice was constructed by using the interatomic potential presented by Andribet et al.[13], and then defects such as a single vacancy, divacancy and vacancy clusters were introduced into the central part of the model lattice. The potential used here covers the change of the chemical bonding due to the strucuture change from sp2 type (graphite) to sp3 type (diamond), which gives reasonable basis to the simulation of defects in graphite. Through the full relaxation of the whole lattice with a defect using the molecular dynamics method the atomic configuration around the defect can be determined. After that positron lifetime calculation was made in the framework of so-called atomic superposition method developed by Puska and Nieminen[14]. The correlation interaction between a positron and electrons was included in the basis of LDA (local density approximation) as given by Boronski and Nieminen[15] and Puska, Seitsonen and Nieminen[16]. The electron wavefunction table for a carbon atom given by Herman and Skillman[17] was used and Schrödinger equation was solved by the method developed by Kimball and Shortley[18]. The positron lifetime was obtained by the inverse of the annihilation rate which is proportional to the product of positron density and electron density accompanied with the

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so-called enhancement factor caused by the correlation effect between a positron and electrons.

3. Result of Calculation

In **Figs. 1** and **2** the atomic configuration around a single vacancy and a divacancy in a basal

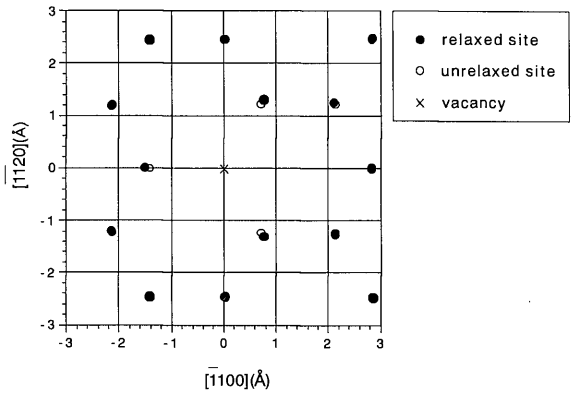

Fig. 1 Atomic configuration around a single vacancy in a basal plane of graphite obtained after the full relaxation by the molecular dynamics method.

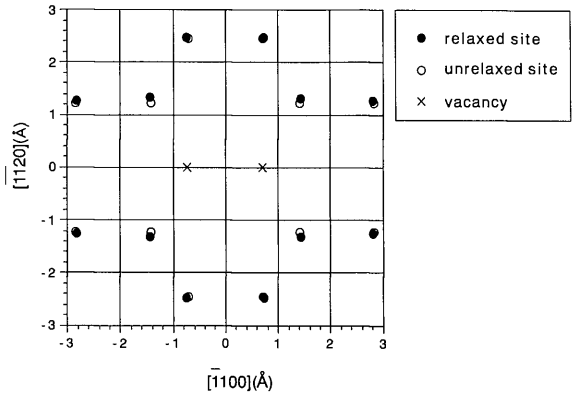

Fig. 2 Atomic configuration around a divacancy in a basal plane of graphite obtained after the full relaxation by the molecular dynamics method.

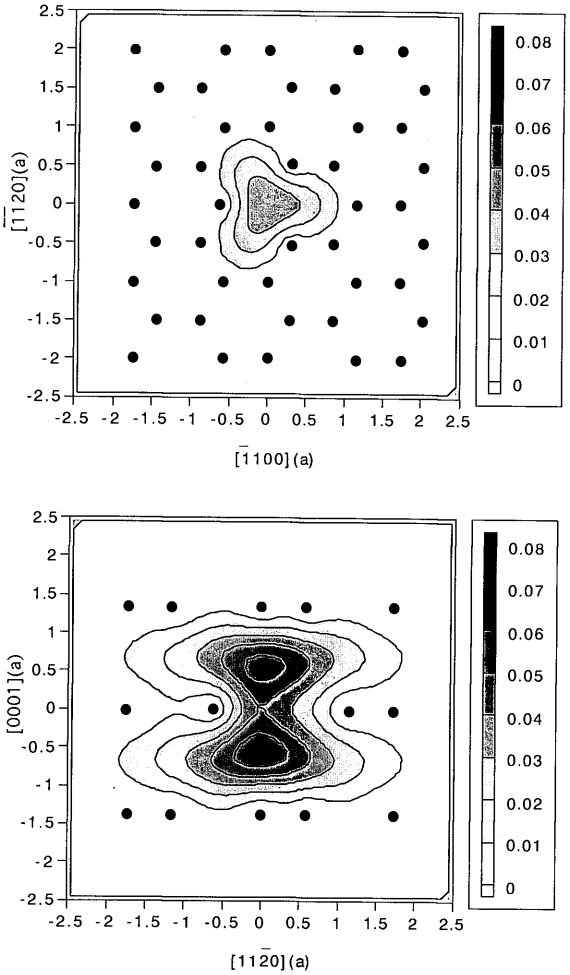

Fig. 3 Wavefunction of a positron trapped at a single vacancy obtained in model graphite lattice (on a basal plane ((0001) plan) (upper), and on an edge ((1120) plan) (bottom)).

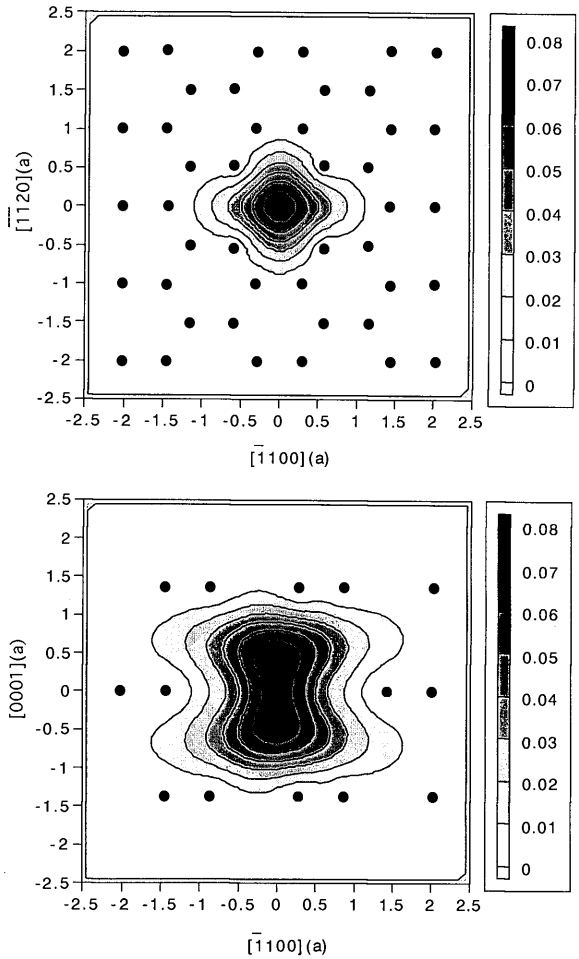

Fig. 4 Wavefunction of a positron trapped at a divacancy obtained in model graphite lattice (on a basal plane ((0001) plan) (upper), and on an edge ((11$\bar{2}$0) plan) (bottom)).

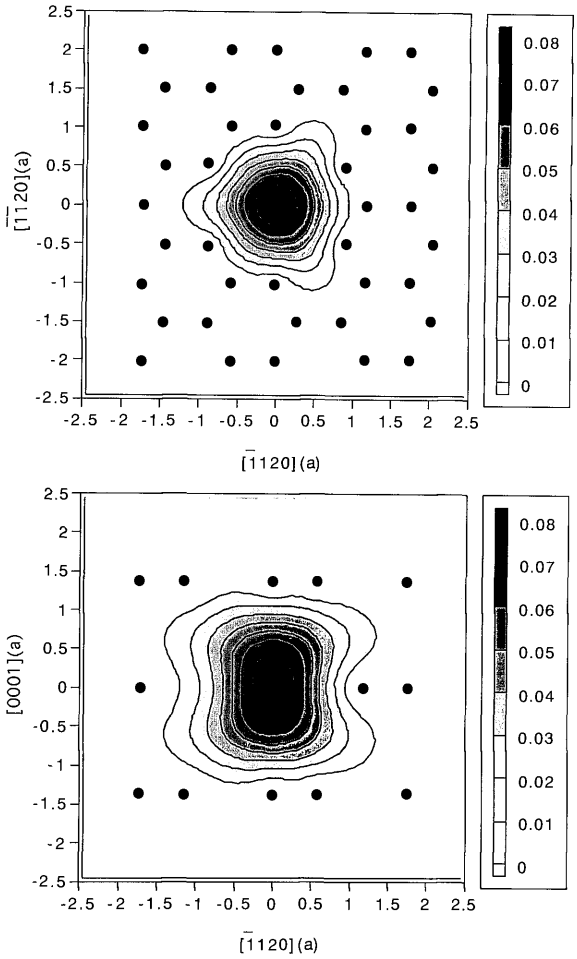

Fig. 5 Wavefunction of a positron trapped at a vacancy cluster V_4 obtained in model graphite lattice (on a basal plane ((0001) plan) (upper), and on an edge ((1120) plan) (bottom)).

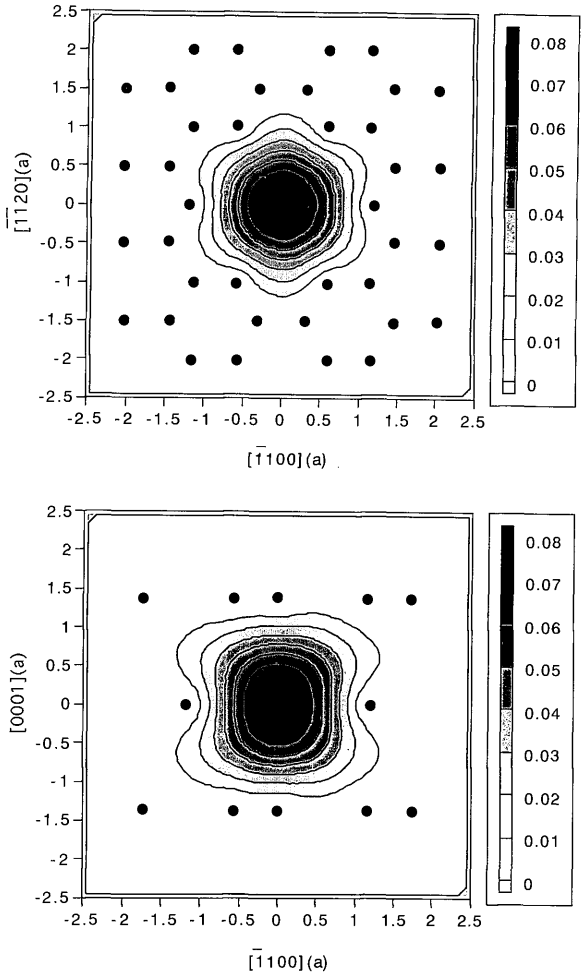

Fig. 6 Wavefunction of a positron trapped at a vacancy cluster V_6 obtained in model graphite lattice (on a basal plane ((0001) plan) (upper), and on an edge plane ((1120) plan) (bottom)).

plane of graphite obtained after the full relaxation by the molecular dynamics method are shown. It is seen that atoms around defects are slightly shifted outwards (~0.1Å), that is, the free volume of a vacancy or a divacancy increases. This tendency keeps to continue even at six vacancy cluster V_6 (vacancy loop V_6) and the another characteristic feature is that the open structure is still kept even after relaxation. The wavefunction of a positron in matrix is not localized and has higher density in the interlayer regions (between basal planes). There are two types of single vacancies, one of which has two carbon atoms just above and below the center of a vacancy and the other has not. A positron is trapped at the either one but the wavefunction is not localized at the central position. The wavefunction of a positron trapped at a vacancy is split into two parts, namely, is extended to the adjacent interlayer regions as seen in **Figs. 3** and **4**. This splitting behavior disappears above V_3 and the positron wavefunction becomes localized at the central position of the clusters as shown in **Figs. 5** and **6**, where positron wavefunctions trapped at V_4 and V_6 are shown, respectively. Calculated values of positron lifetimes for a vacancy and vacancy clusters are shown in **table 1** and **Fig. 7**, where values for both unrelaxed and relaxed structure are listed and the increasing feature of positron lifetime with increasing number of vacancies in one cluster is seen.

4. Comparison with Experimental Results

Positron lifetime 208psec was obtained for the matrix of HOPG (highly oriented pyrolytic

Table 1 Calculated positron lifetimes at a vacancy and vacancy clusters in graphite.

Defects	Positron Lifetime [psec]	
	Unrelaxed	Relaxed
Matrix	204	—
V_1	216	222
V_2	242	256
V_3	277	296
V_4	329	352
V_6	434	458

graphite) by the present authors, and 233psec for a single vacancy in the electron irradiated (2.5 MeV electrons at room temperature[19]) HOPG. Calculated results for these, i.e., 204 psec and 222 psec are considered to be in good agreement with these experimental results. In **Fig. 8** is shown the result of the isochronal annealing experiment above room temperature due to the positron annihilation lifetime measurement for HOPG irradiated by 28 MeV electrons at 77K to a dose of $4.5 \times 10^{18} e/cm^2$ at KURRI Linac. Since the electron energy is high, the second lifetime component at room temperature is higher than 233 psec (for a single vacancy), suggesting that small vacancy clusters are already existing at room temperature without migration of vacancies. The isochronal annealing behavior in **Fig. 8** can be roughly divided into two regions, that is, region I (below 1200℃) and region II (above 1200℃). It can be considered that the increase of positron lifetime in each region corresponds to formation process of interstitial clusters and vacancy clusters, respectively. Vacancies start to migrate at high temperature region between these two stages, which causes the decrease of lifetime through arriving of vacancies at interstitial clusters, resulting in the shrinkage of them. In region II not only growth of vacancy clusters but also growth of interstitial clusters continuing from region I might be included. The longest lifetime is near 400 psec as seen in region II in **Fig. 8**, and this might correspond to V_4 and V_6 in **table 1**.

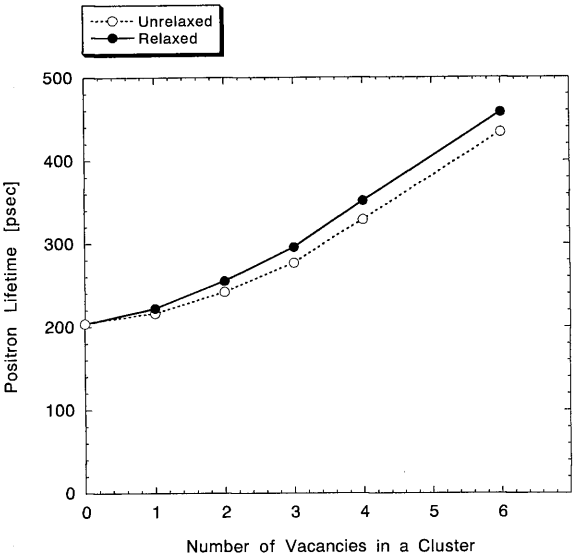

Fig. 7 Caculated positron lifetimes at a vacancy and cluters in graphite as a function of number of vacancies in a cluster.

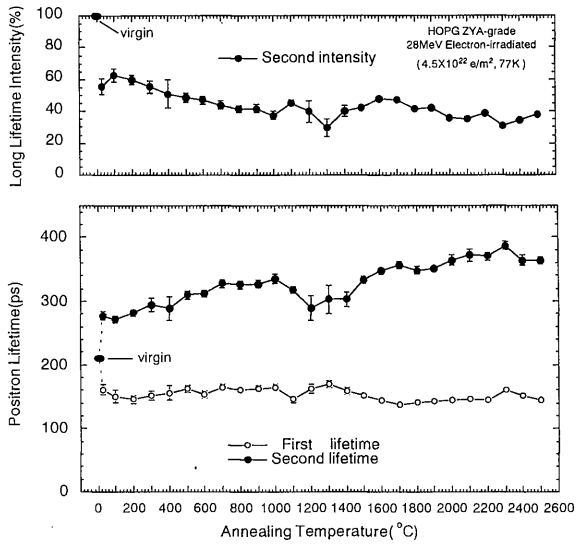

Fig. 8 Result of the isochronal annealing experiment adove room temperature due to the positron annihilation lifetime measurement for HOPG irradiated by 28 Mev electrons at 77K to a dose of $4 \times 10^{18} e/cm^2$ at KURRI Linac.

5. Conclusion

Positron lifetime calculation in the framework of the atomic superposition method was made for the matrix, a single vacancy and vacancy clusters in the model graphite lattice constructed by using the potential presented by Andribet et al., and relaxed by molecular dynamics method. Reasonalbe agreement with experiments was obtained for the matrix and a single vacancy and vacancy clusters.

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