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Positron lifetime calculation for defects and defect clusters in graphite

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Abstract

Calculations of positron lifetime have been made for vacancy type defects in graphite and compared with experimental results. Defect structures were obtained in a model graphite lattice after including relaxation of whole lattice as determined by the molecular dynamics method, where the interatomic potential given by Pablo Andribet, Dominguez-Vazguez, Mari Carmen Perez-Martin, Alonso, Jimenez-Rodriguez [Nucl. Instrum. and Meth. 115 (1996) 501] was used. For the defect structures obtained via lattice relaxation positron lifetime was calculated under the so-called atomic superposition method. Positron lifetimes 204 and 222 ps were obtained for the graphite matrix and a single vacancy, respectively, which can be compared with the experimental results 208 and 233 ps. For planar vacancy clusters, e.g., vacancy loops, lifetime calculation was also made and indicated that lifetime increases with the number of vacancies in a cluster. This is consistent with the experimental result in the region of higher annealing temperature (above 1200°C), where the increase of positron lifetime is seen, probably corresponding to the clustering of mobile vacancies.

1. Introduction

It is well known that graphite has recently become one of the important candidates for the plasma facing materials in a fusion reactor, because of its excellent performance at high temperature region, such as high thermal conductivity and low Z character [1]. Hydrogen recycling in graphite is a severe problem for the plasma confinement in the fusion reactor and is tightly connected to the defect behaviors in surface region of graphite. Hence, defect properties in graphite must be investigated more in detail [2–10]. On the other hand, positron annihilation technique is very sensitive to the presence of vacancy type defects and the calculation method of positron lifetime has recently advanced to a large extent, 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 comparison with experimental results will also be made.

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. Graphite crystal consists of two kinds of bonding, that is, covalent bonding in a basal plane, and van der Waals bonding between two adjacent basal planes. The potential used here covers the change of the chemical bonding due to the structure 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 this process positron lifetime calculation was made in the framework of so-called atomic superposition method

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developed by Puska and Nieminen [14]. The correlation interaction between a positron and electrons was included in the basis of local density approximation (LDA) as given by Boronski and Nieminen [15] and Puska et al. [16]. The electron wave function table for a carbon atom given by Herman and Skillman [17] was used and Schrödinger equation was numerically 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 by the so-called enhancement factor arising from the correlation effect between a positron and electrons.

3. Result of calculation

Figs. 1 and 2 show the atomic configuration around a single vacancy and that around a divacancy in a basal plane of graphite, respectively. These were obtained after the full relaxation of the whole lattice with a defect at the center by using the molecular dynamics method. It is clearly seen that atoms around defects are slightly shifted outwards (~0.1 Å), resulting in the increase of the free volume of a vacancy or a divacancy. This tendency continues even at nine vacancy cluster V_9 (vacancy loop V_9) and another characteristic feature is that open structure of these vacancy clusters still remains even after relaxation, which is quite different from the case of usual metals where planar vacancy clusters usually have collapsed structure.

Although the wave function of a positron in matrix basically has a delocalized nature, it has higher density in the interlayer regions (between two adjacent 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

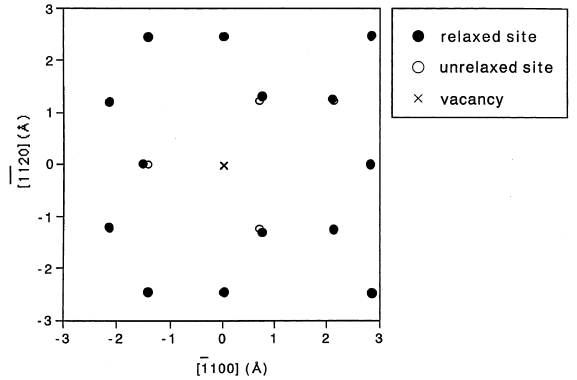

Fig. 1. Atomic configuration around a single vacancy in a basal plane of graphite obtained after the full relaxation by the molecular dynamics method. (○ before relaxation, ● after relaxation, × vacancy.)

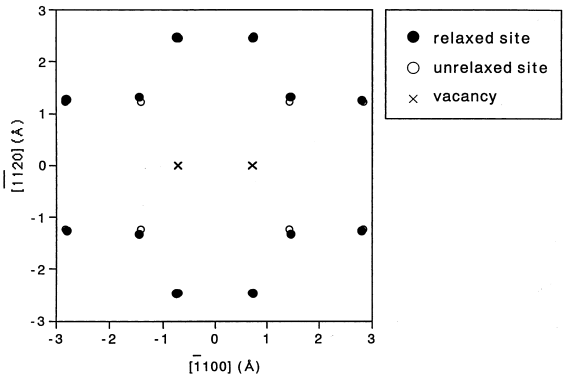

Fig. 2. Atomic configuration around a divacancy in a basal plane of graphite obtained after the full relaxation by the molecular dynamics method. (○ before relaxation, ● after relaxation, × vacancy.)

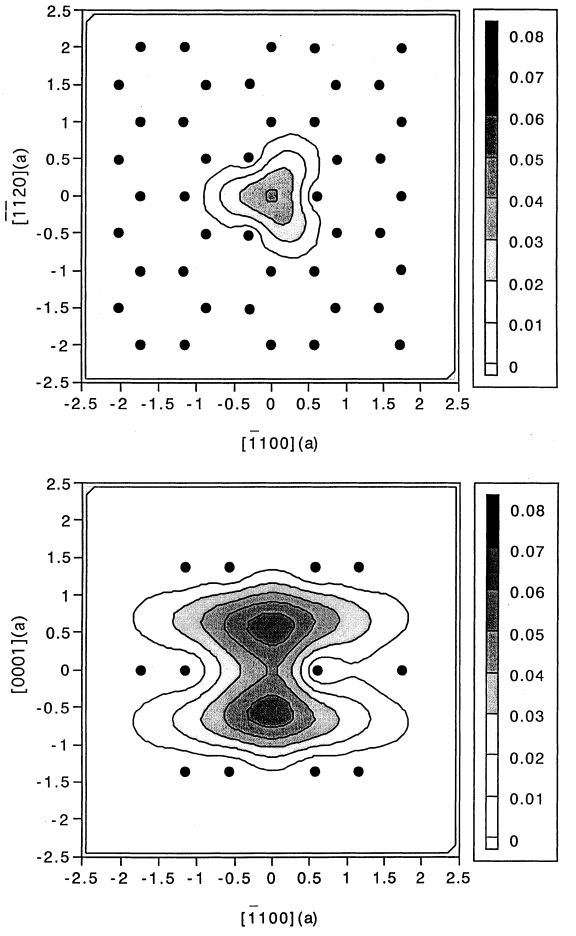

Fig. 3. Wave function of a positron trapped at a single vacancy obtained in model graphite lattice (on a basal plane ((0 0 0 1) plane) (upper), and on an edge plane ((1 1 2 0) plane) (bottom)).

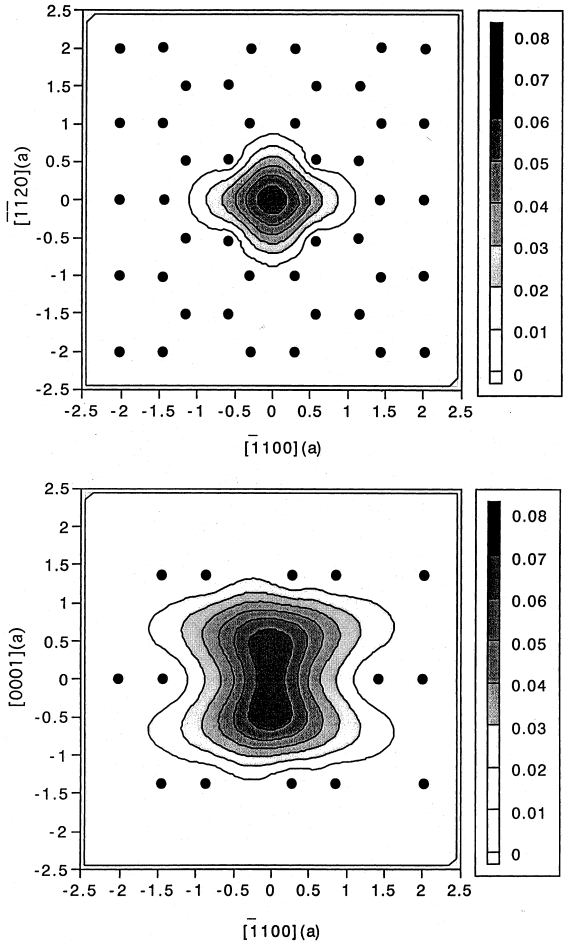

Fig. 4. Wave function of a positron trapped at a divacancy obtained in model graphite lattice (on a basal plane ((0 0 0 1) plane) (upper), and on an edge plane ((1 1 2 0) plane) (bottom)).

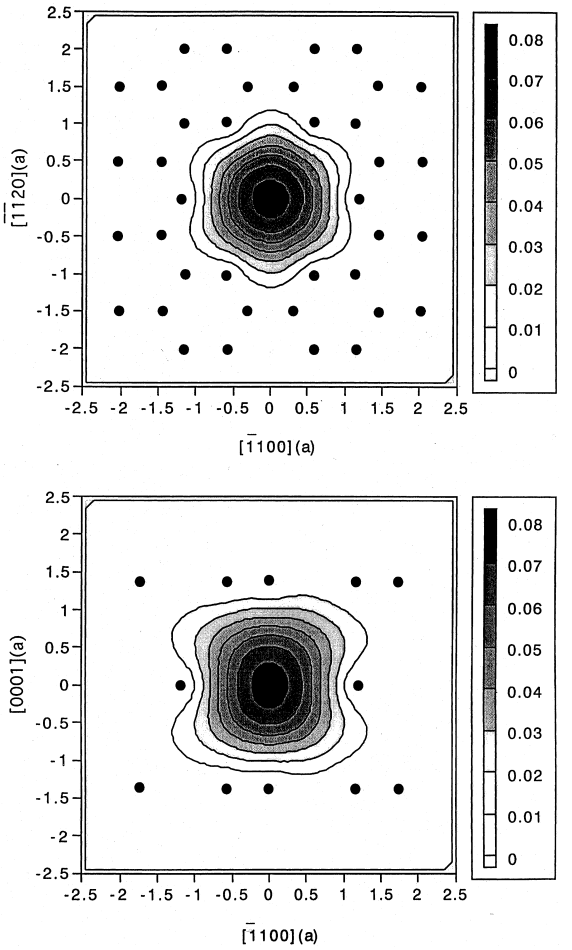

Fig. 5. Wave function of a positron trapped at a vacancy cluster V_6 obtained in model graphite lattice (on a basal plane ((0 0 0 1) plane) (upper), and on an edge plane ((1 1 2 0) plane) (bottom)).

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

Defect type	Positron lifetime (ps)		
	Calculation		Experimental
	Unrelaxed	Relaxed	
Matrix	204	–	208
V_1	216	222	233
V_2	242	256	–
V_4	246	282	–
V_6	319	352	(300–400)
V_9	370	395	–

trapped at the either one but the wave function is not localized at the central position of a vacancy. The wave function of a positron trapped at a vacancy or at a divacany is split into two parts, namely, is extended to the adjacent interlayer regions as seen in Figs. 3 and 4, respectively. This splitting behavior disappears above V_3 and the positron wave function becomes localized at the central position of the clusters as shown in Figs. 5 and 6, where positron wave functions trapped at V_6 and V_9 are shown, respectively. Calculated values of positron lifetimes for a vacancy and vacancy clusters are shown in Table 1, where values for both unrelaxed and relaxed structure are listed and the increasing tendency of positron lifetime with increasing number of vacancies in one cluster is clearly seen.

4. Comparison with experimental results

From the experiments so far performed, it is known that positron lifetime for the matrix of highly oriented

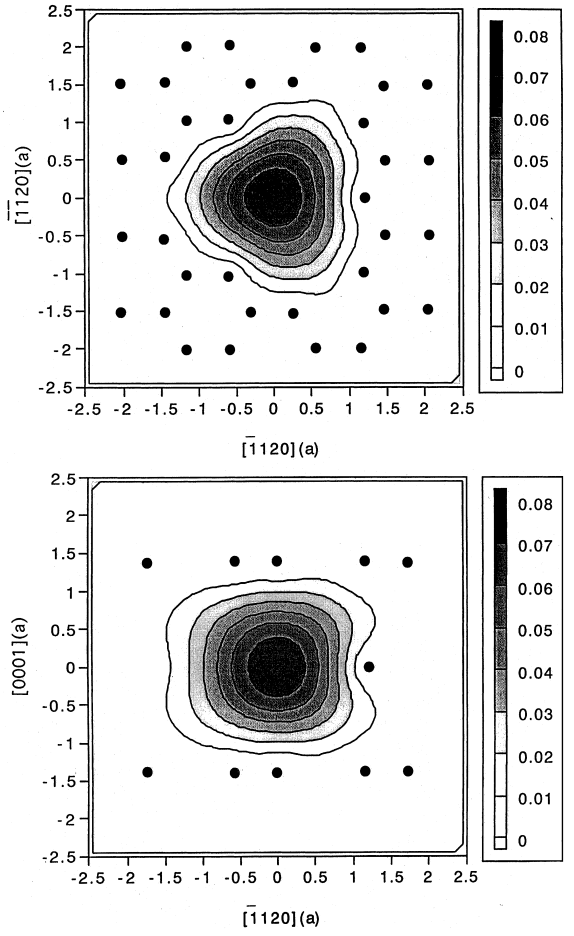

Fig. 6. Wave function of a positron trapped at a vacancy cluster V_9 obtained in model graphite lattice (on a basal plane ((0 0 0 1) plane) (upper), and on an edge plane ((1 1 2 0) plane) (bottom)).

pyrolytic graphite (HOPG) is 208 ps (present authors), and that for a single vacancy is 233 ps, which was obtained from the measurement for the HOPG irradiated with 2.5 MeV electrons [19]. Positron lifetimes obtained from the calculation for matrix and a vacancy are 204 and 222 ps, respectively. These are considered to be in good agreement with experimental results. In Fig. 7, the result of the isochronal annealing experiment above room temperature made by the positron annihilation lifetime measurement for HOPG irradiated by 28 MeV electrons is shown at 77 K to a dose of 4.5×10^{18} e/cm^2 at KURRI Linac. Since the electron energy is high, the second lifetime component at room temperature is higher than 233 ps (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. 7 can be roughly

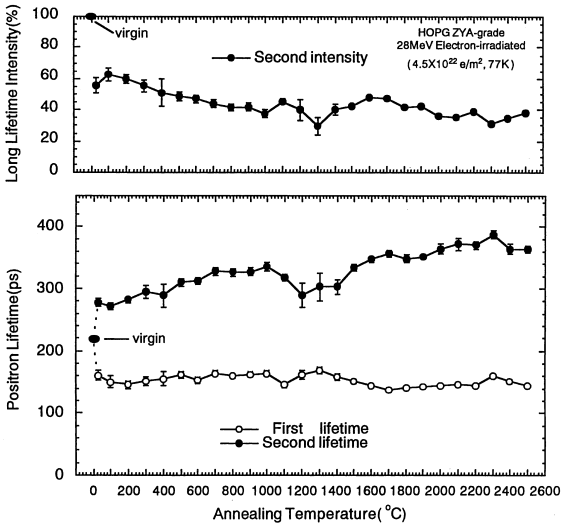

Fig. 7. Result of the isochronal annealing experiment above room temperature made by the positron annihilation lifetime measurement for HOPG irradiated by 28 MeV electrons at 77 K to a dose of 4×10^{18} e/cm^2 at KURRI Linac.

divided into two regions, that is, region I (below 1200°C) and region II (above 1200°C).

It is reported that migration energies of an interstitial atom and a vacancy in graphite are 0.1 and 3.1 eV in a basal plane, respectively [5]. It can be therefore considered that the increase of positron lifetime in region I and II corresponds to growth process of interstitial clusters (loops) and vacancy clusters (loops), respectively. It is reasonably considered that the increase of positron lifetime for the growth of interstitial clusters comes from the open structure around the peripheral position of a loop arising from the weak van der Waals bonding. This open structure is considered to be firmly established with increasing loop size. On this point more detailed calculations will be required. Vacancies start to migrate around 1200°C between these two stages, which cause the decrease of lifetime through arriving of vacancies at interstitial clusters, resulting in their shrinkage. 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 ps as seen in region II in Fig. 7, and this might correspond to V_6 and V_9 in Table 1.

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. [13], and relaxed by molecular dynamics method. Reasonable agreement with experiments was obtained for the matrix and a single vacancy and vacancy clusters.

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