Positron Lifetime Calculation of Vacancy Clusters in Ta Containing H and He

Research Reactor Institute
Kyoto University
Q. XU, J. Zhang
Bulgarian Academy of Sciences
E. Popov, T. Troev
Paul Scherrer Institut
Y. Dai

Background

Desirable properties of a target material for spallation source with high power

- ✓ High atomic number
- ✓ High density
- ✓ High melting point (for solid target)
- ✓ High thermal conductivity
- ✓ Low corrosion
- ✓ Resistance to radiation damage
- ✓ Low absorption for neutron
- ✓ Good availability and low price

W and Ta (and their alloys) are usually considered materials for solid targets.
Corrosion resistance of W is poor in the cooling water.
Ta-claddings can solve the problem of the corrosion, therefore, Ta-cladded W is considered to be the candidated target for spallation neutron source.

Productions of H and He induced by spallation neutrons are higher than those induced by fission and fusion neutrons.

Damage and He production in 316 steel

Irr. environment	Damage (dpa/y)	He (appm/y)
Fusion neutrons	32.1	465
HFIR Fission neutrons	36	2187
EBR-II Fission neutrons	39.2	15.1
SNS SB*	34	3000

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Object: investigation of microstructural evolution in Ta irradiated by spallation neutrons

Formation of vacancy clusters (containing H or He or H+He)

Calculation Method of Positron Lifetime Based on DFT

$$\{-\nabla^2 + V_{eff}[n(r), r]\}\varphi_i(r) = \varepsilon_i \varphi_i(r)$$

wave function

eigen values

effective potential $V_{eff}[n(r), r] = \emptyset(r) + V_{xc}[n(r)]$

electrostatic potential

Exchange correlation potential

electron density $n(r) = \sum_i |\varphi_i(r)|^2$

Positron wave function is obtained by a method similar to above

$$\{-\nabla^2 + V_+[n(r), r]\}\varphi_+(r) = \varepsilon_+ \varphi_+(r)$$

Positron lifetime or annihilation rate in an inhomogeneous electron gas:

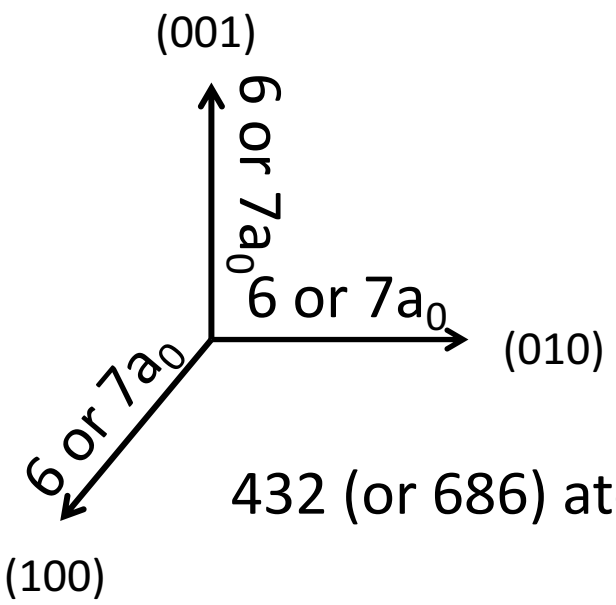
$$\frac{1}{\tau} = \lambda = \int dr |\varphi_+(r)|^2 (\Gamma_V[n_v(r)] + \Gamma_{in}[n_{in}(r)])$$

valence electron

core electron

Calculation Method of Positron Lifetime (cont.)

Cubic Cell



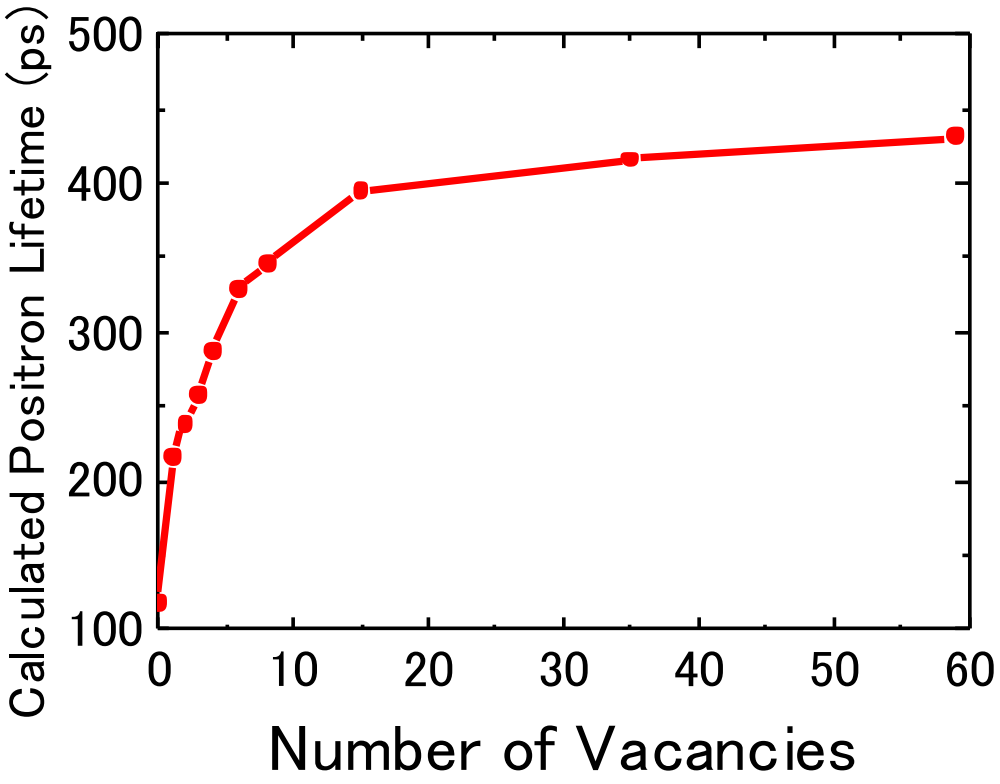
periodic boundary conditions

Time step: 0.0025 ps

432 (or 686) atoms

Lattice Constant of Ta: a_0=0.28665 nm

Lifetimes of Vacancy and Vacancy Clusters

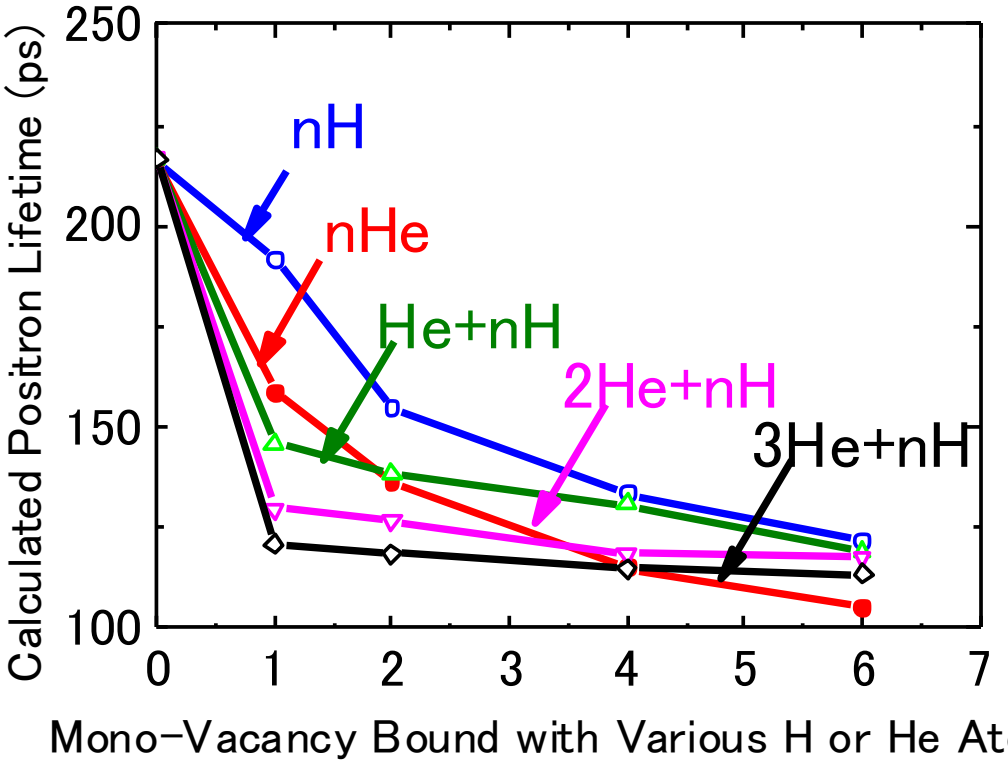


matrix: 118.3 ps

mono-V: 216.5 ps

Positron lifetime of vacancy-cluster increases with increasing the number of vacancies in cluster, and tends to saturate around the values of 400 ps for cluster containing 15 vacancies.

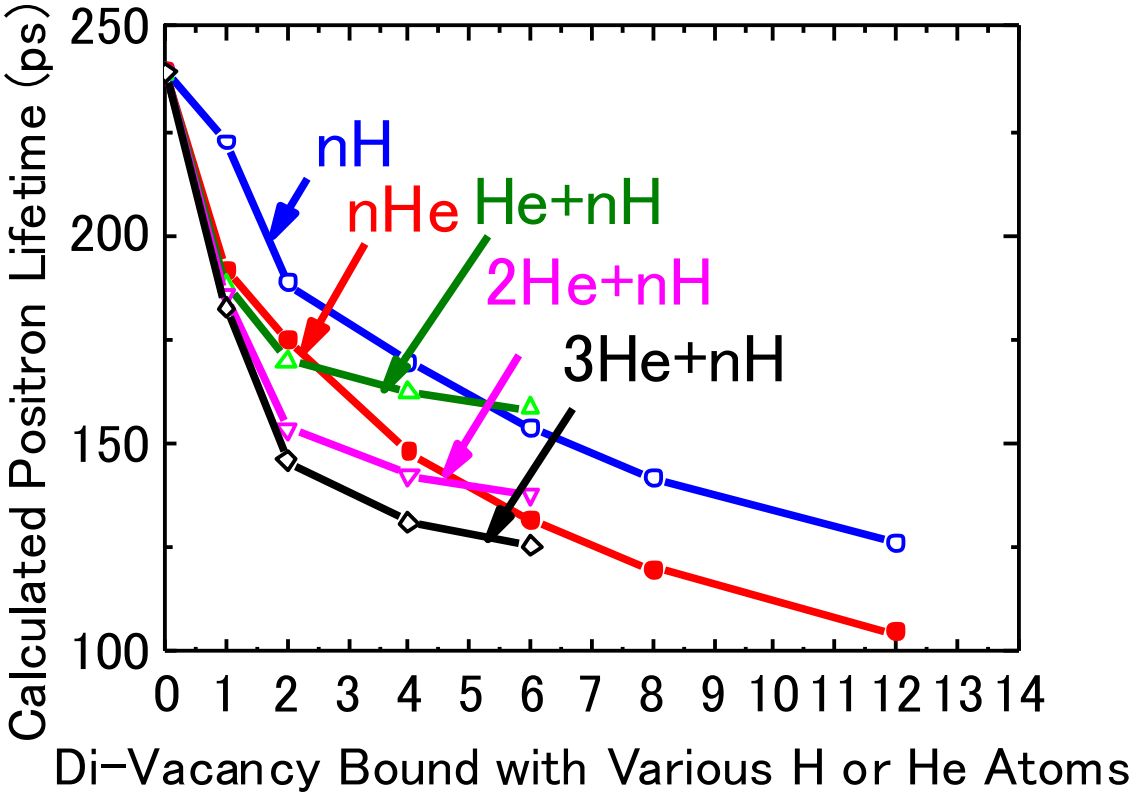
Effects of H and He on Lifetime of 1V



1V: 216.5 ps
1V+1H: 191.7 ps
1V+6H: 121.7 ps
1V+1He: 158.6 ps
1V+6He: 105.2 ps
1V+1H+1He: 145.8 ps

- The positron lifetime of mono-vacancy decreases to 191.7 ps after absorption of 1H, and it decreases gradually with increasing the number of H atoms.
- The positron lifetime of mono-vacancy decreases to 158.6 ps after absorption of 1He, and it decreases gradually with increasing the number of He atoms.
- The decrease in lifetime is predominant with increasing the number of He atoms.
- The positron lifetime of mono-vacancy decreases after absorption of both H and He.

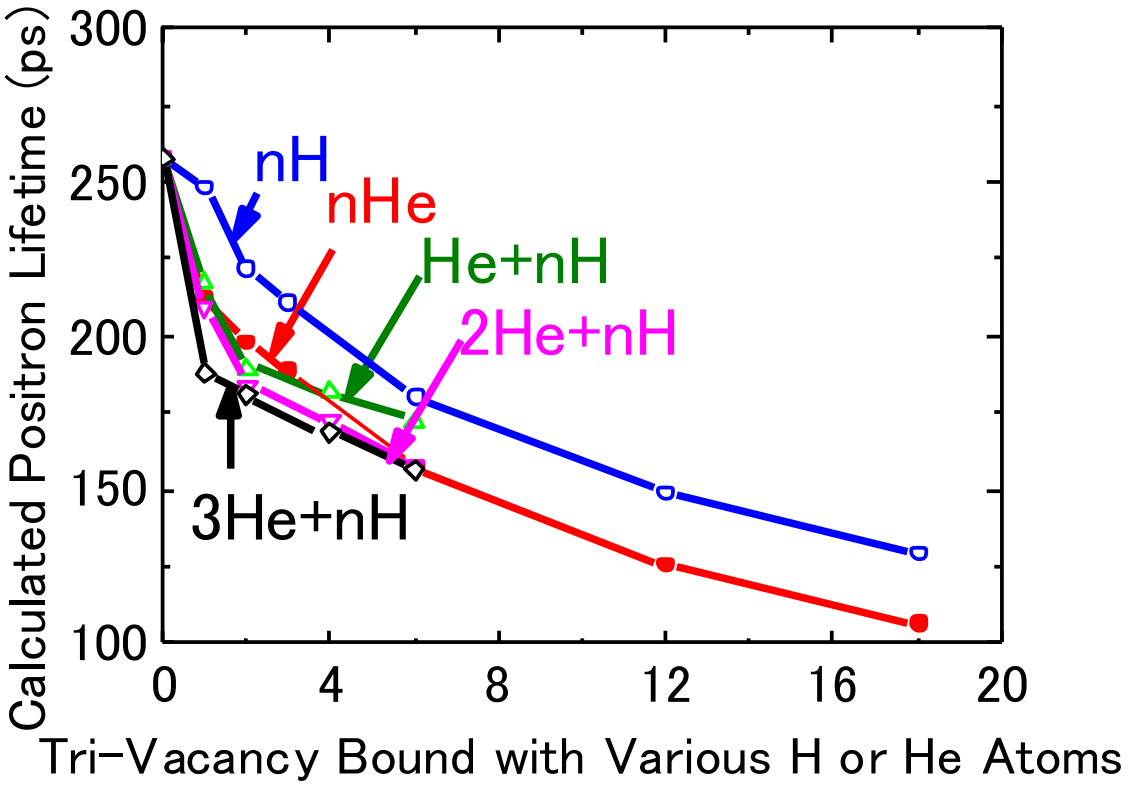
Effects of H and He on Lifetime of 2V



2V: 239.1 ps
2V+1H: 222.2 ps
2V+6H: 153.3 ps
2V+1He: 191.3 ps
2V+6He: 131.3 ps
3V+1H+1He: 187.7 ps

- The positron lifetime of di-vacancy decreased from 239.1 ps to 222.2 and 191.3 ps after absorption of 1H and 1He.
- The decrease in positron lifetime of vacancy clusters is more prominent in He absorption than that in H absorption.

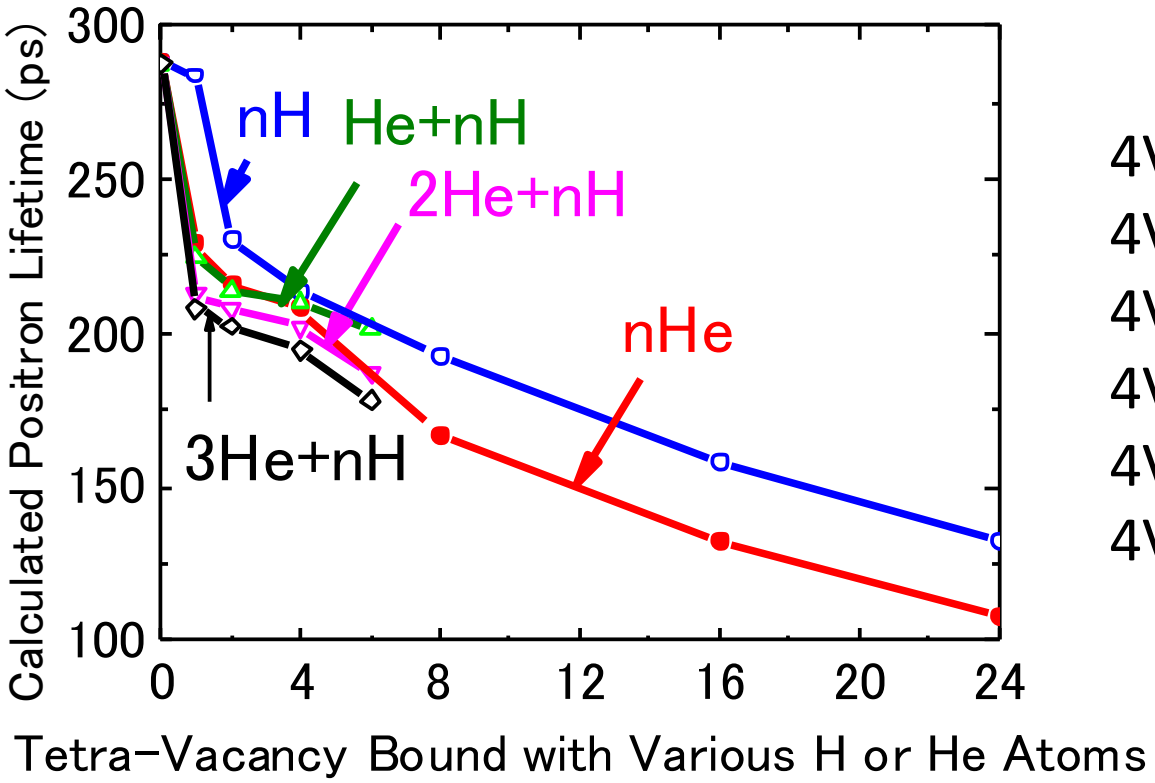
Effects of H and He on Lifetime of 3V



3V: 258.0 ps
3V+1H: 248.2 ps
3V+6H: 180.2 ps
3V+1He: 212.1 ps
3V+6He: 157.3 ps
3V+1H+1He: 217.0 ps

- The positron lifetime of tri-vacancy decreased from 258. ps to 248.2 and 212.1 ps after absorption of 1H and 1He.
- The decrease in positron lifetime of vacancy clusters is more prominent in He absorption than that in H absorption.

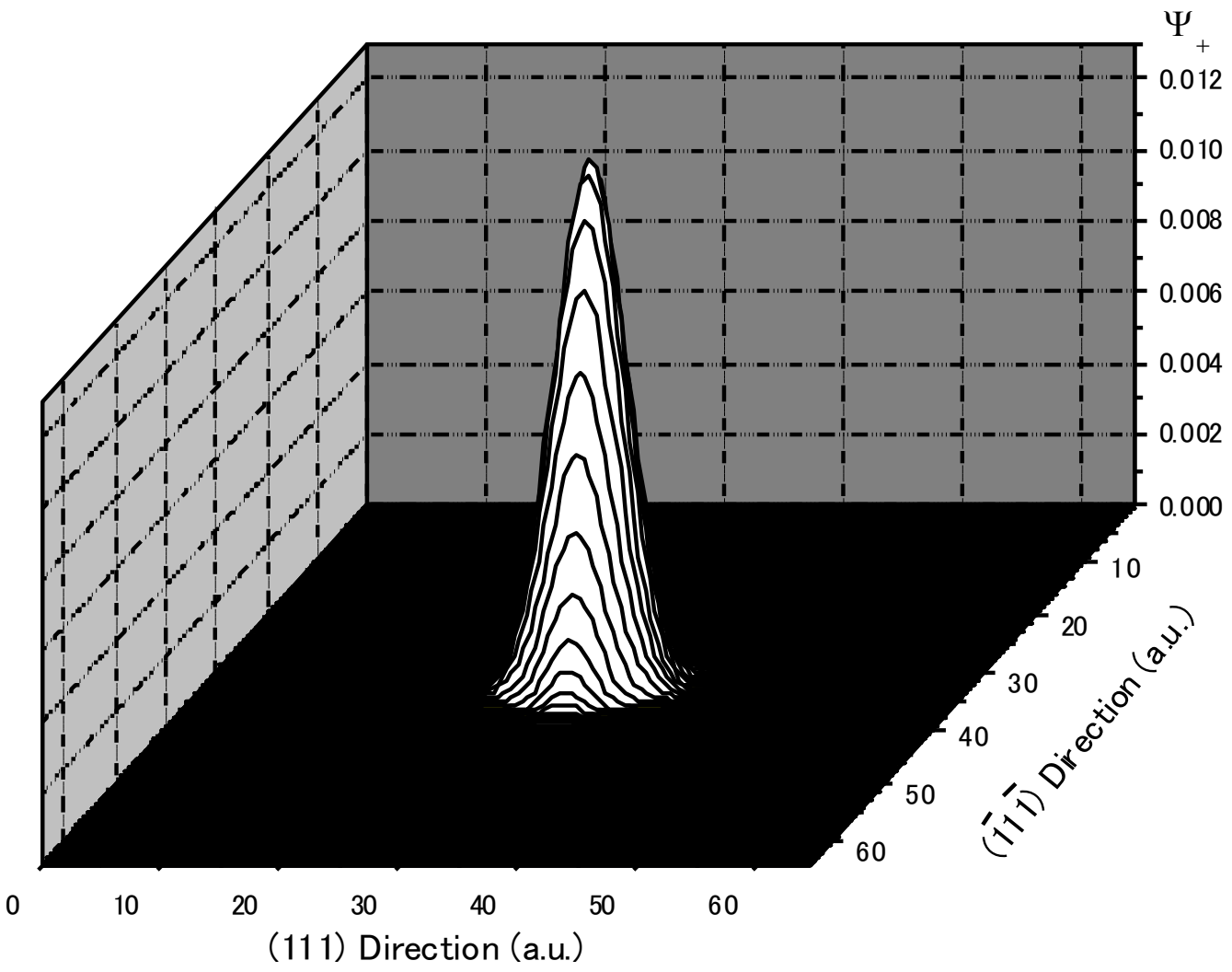
Effects of H and He on Lifetime of 4V



4V: 287.6 ps
4V+1H: 283.8 ps
4V+8H: 192.4 ps
4V+1He: 228.9 ps
4V+8He: 166.5 ps
4V+1H+1He: 225.3 ps

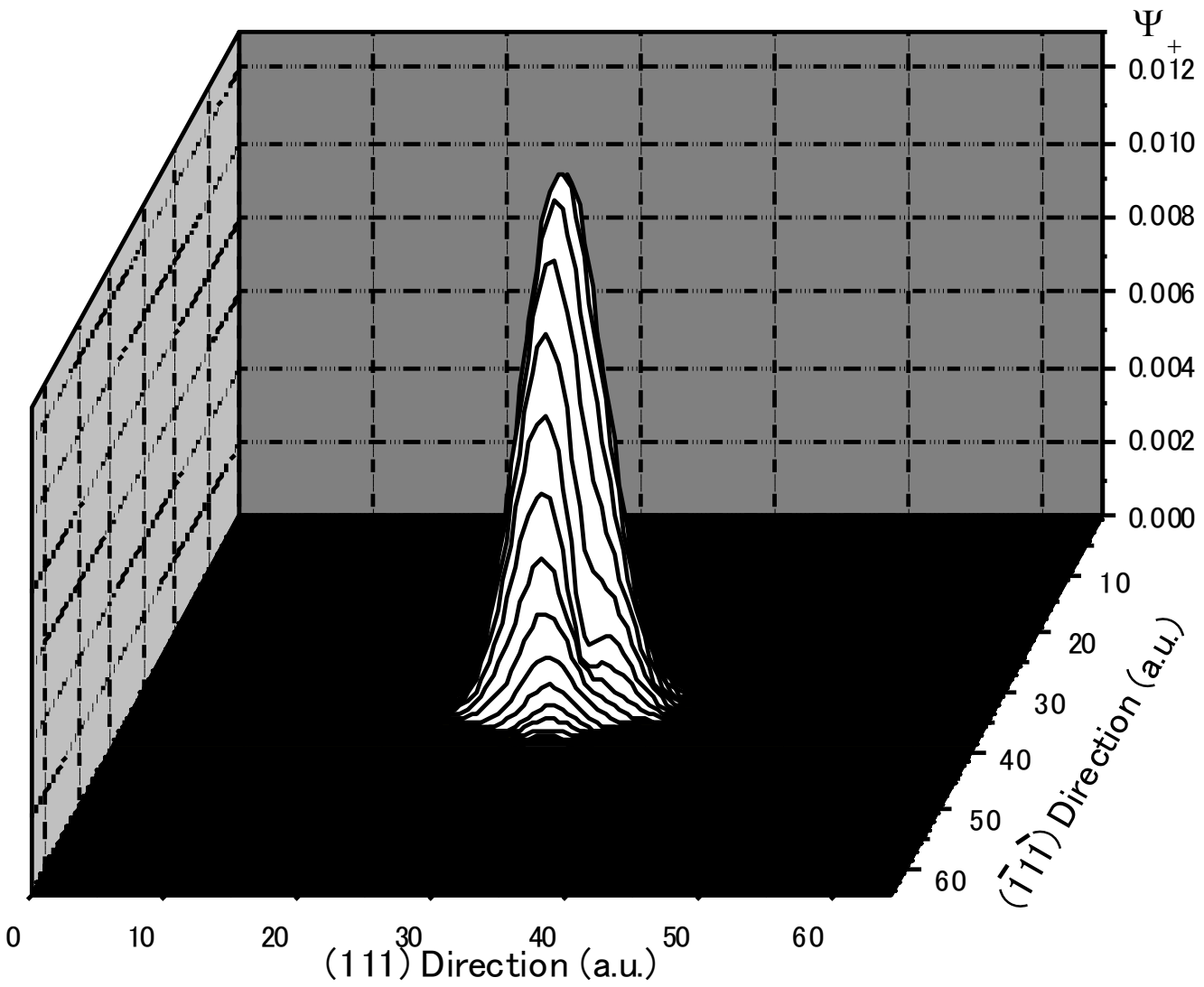
- The positron lifetime of di-vacancy decreased from 287.6 ps to 283.8 and 228.9 ps after absorption of 1H and 1He.
- The decrease in positron lifetime of vacancy clusters is more prominent in He absorption than that in H absorption.

Localized Positron Wave Function in 1V



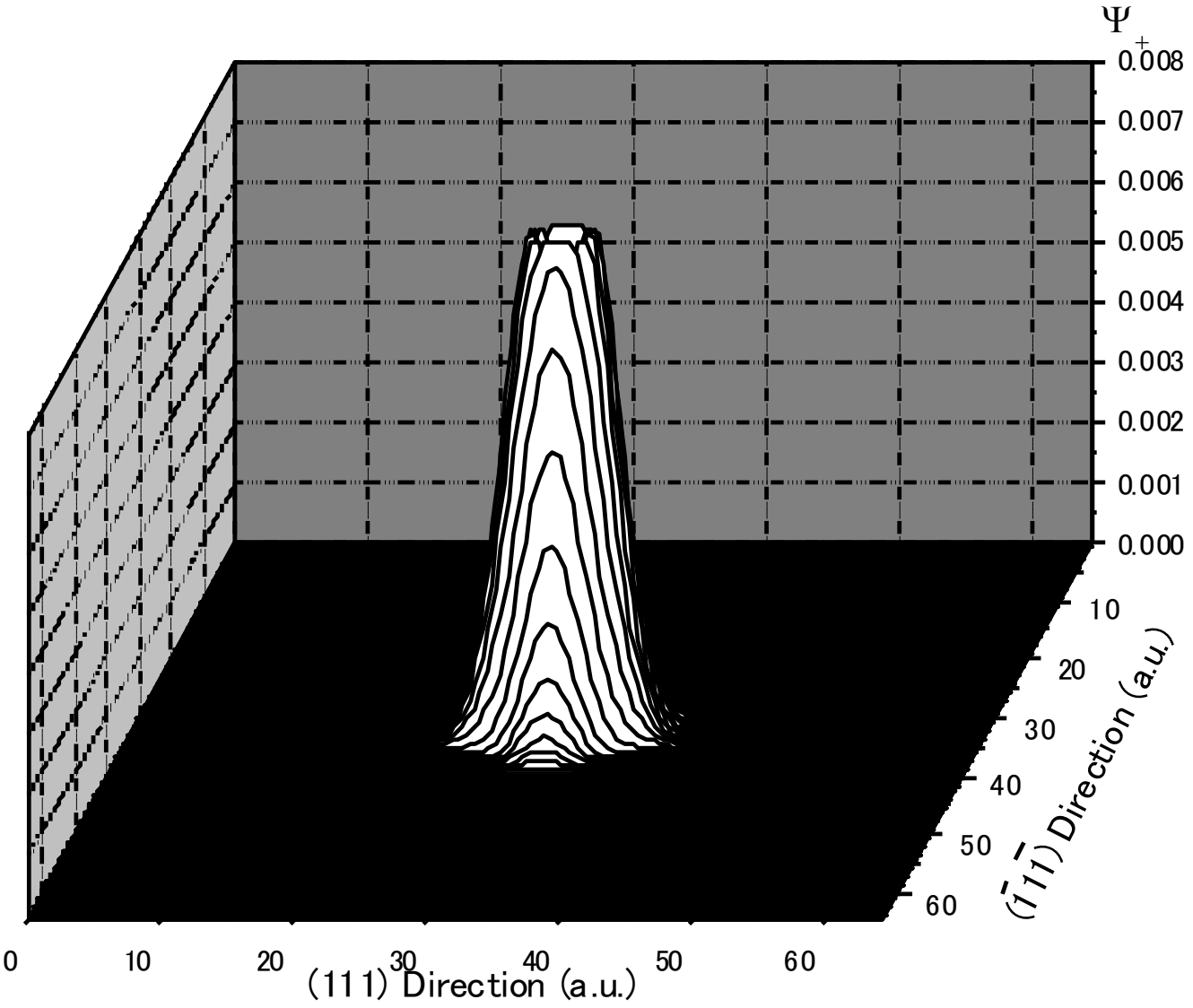
It is clear that positron is localized at the center of the mono-vacancy.

Localized Positron Wave Function in 1V+1H



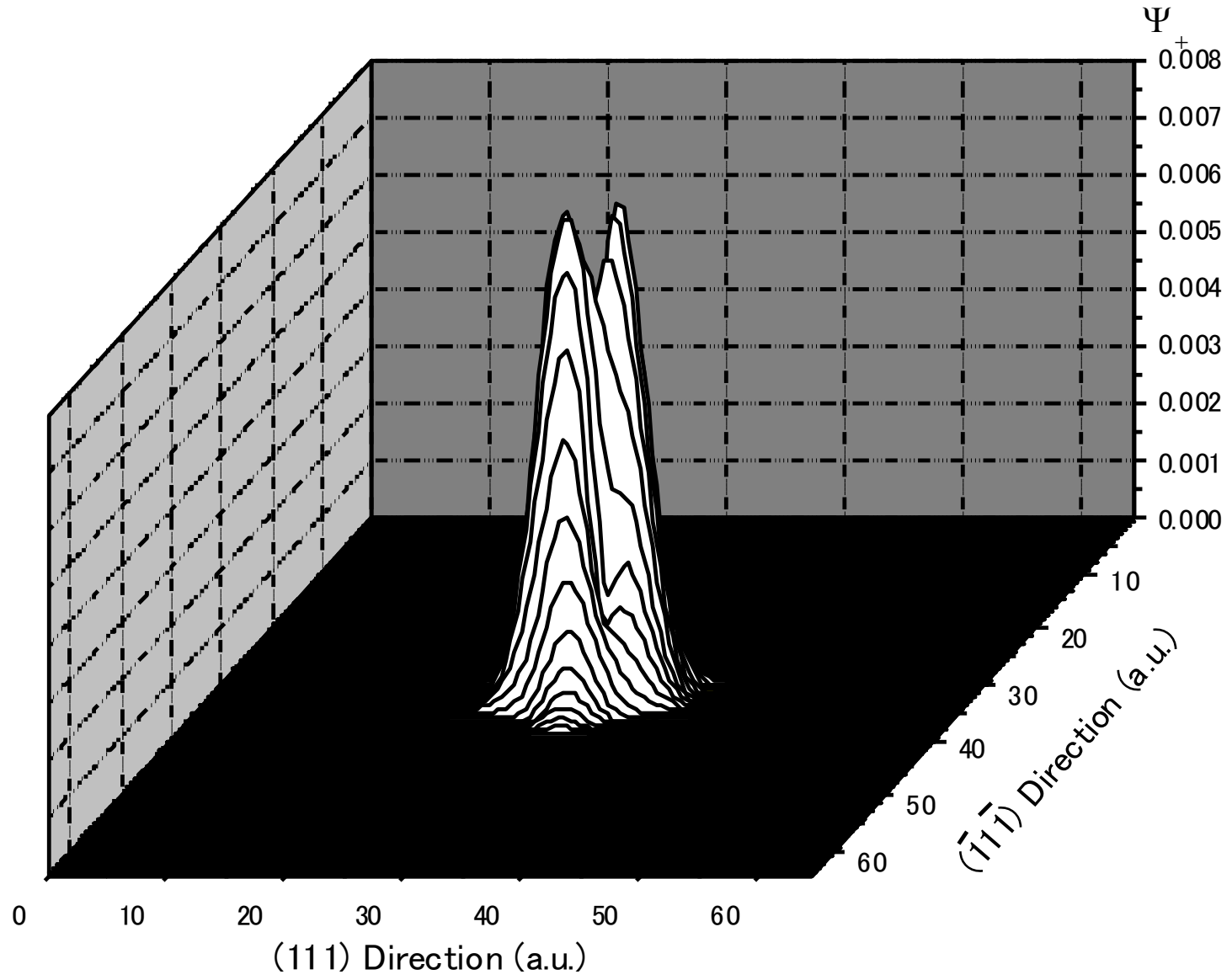
The positron density distribution in 1V+1H is the same as that in 1V.

Localized Positron Wave Function in 1V+1He



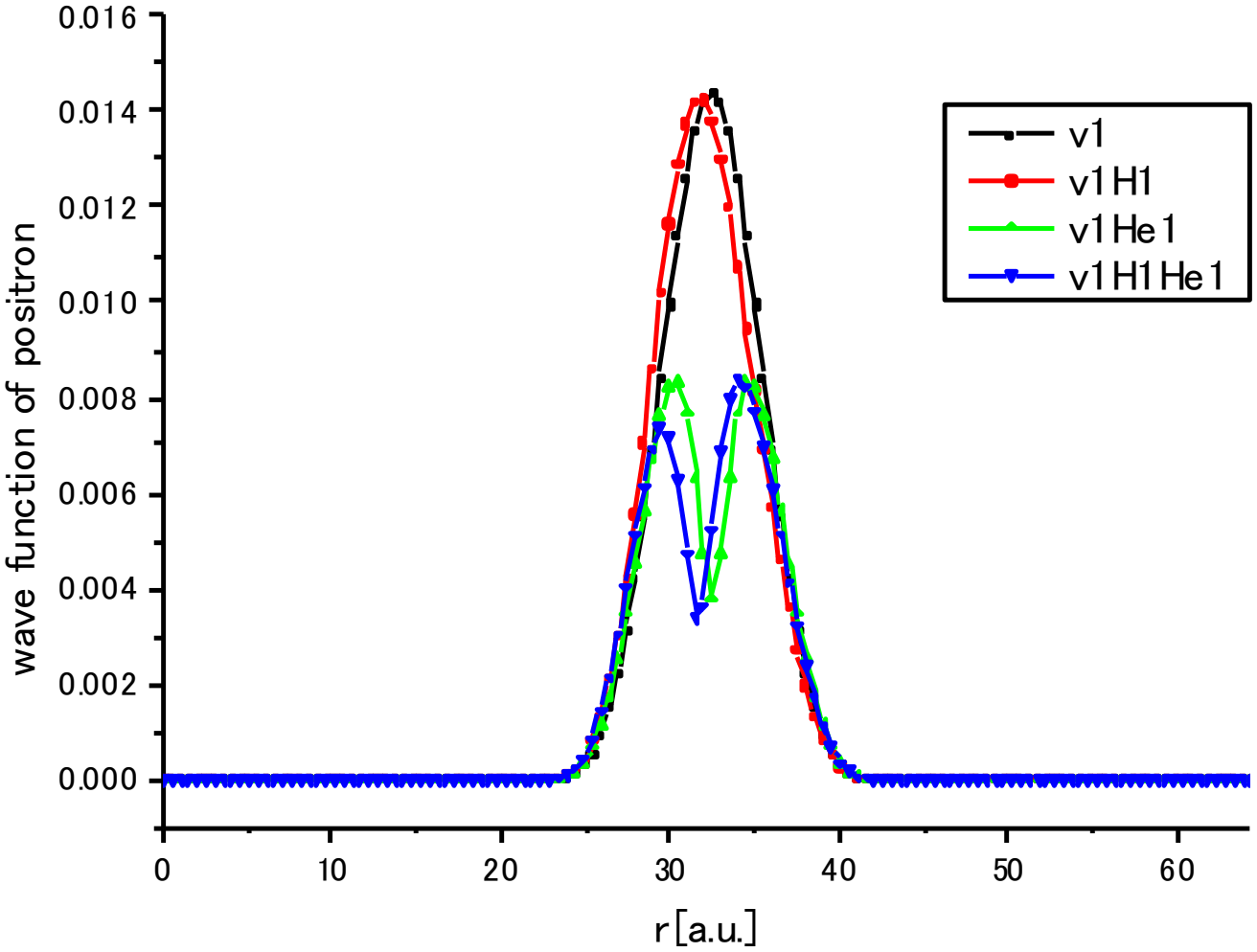
Positron is not localized at the center of the 1V when it contains a He atom

Localized Positron Wave Function in 1V+1H+1He

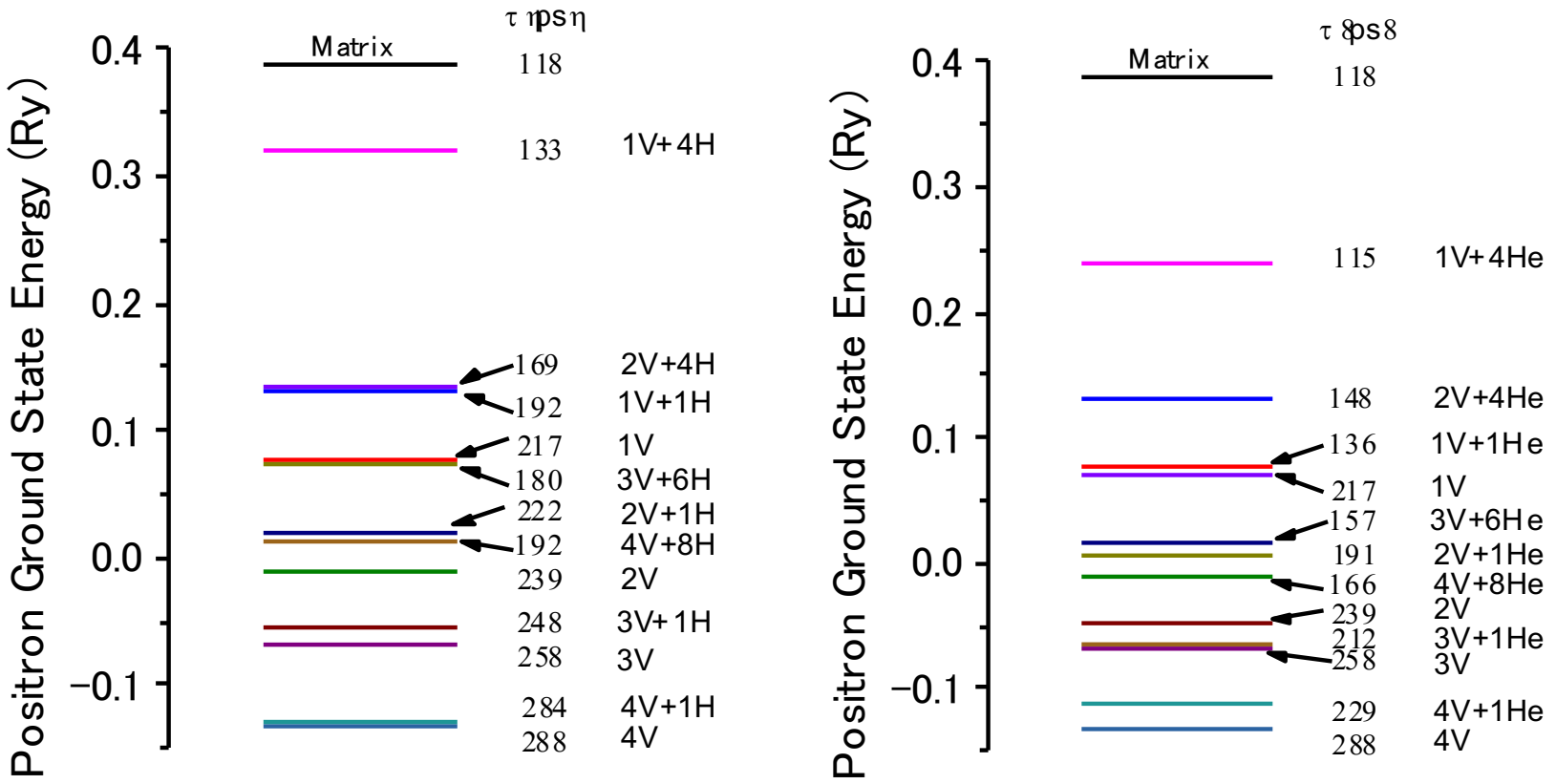


The positron density distribution in 1V+1H+1He is the same as that in 1V+1He.

Comparison of Localized Positron Wave Functions



Comparison of Positron Ground-State Energy of V with H and He



- Positron lifetime decreased but the positron ground-state energy increased when the vacancy clusters bounded H or He atoms.
- Effect of H increasing positron ground-state energy of vacancy cluster is more prominent than that of He.

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Associated strongly with electron distribution around the vacancy cluster

Conclusions

To estimate the interaction between vacancy or vacancies clusters and H, He atoms in Ta, the positron lifetime calculations of V-H, V-He and V-H-He clusters were carried out using quantum-mechanical electronic-structure based on the DFT.

- The lifetime of perfect Ta is 118.3 ps, and it is 216.5 ps when single vacancies are exists.
- The lifetime increases with increasing in number of vacancy in vacancy cluster, and it decreases with increasing in number of H or He or H+He atoms.
- It is clear that the effect of He on lifetime change is larger than that of H.