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The Irradiation Effect of a Simultaneous Laser and Electron Dual-beam on Void Formation

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Randomly distributed lattice point defects such as supersaturated vacancies (SVs) and Frenkel-pairs (FPs, an interstitial and a vacancy) can be simultaneously introduced into the crystal by energetic beam irradiation in outer space and/or nuclear reactors, but their behavior has not been fully understood. Using a high-voltage electron microscope equipped with a laser (laser-HVEM), we show the striking effects of simultaneous laser-electron (photon-electron) dual-beam irradiation on void formation. Our results reveal that during laser-electron sequential irradiation, pre-laser irradiation enhanced void nucleation and subsequent electron irradiation enhanced void growth. However, the laser-electron dual-beam irradiation was analyzed to depress void swelling remarkably because the recombination of SVs and interstitials was enhanced. The results provide insight into the mechanism underlying the dual-beam radiation-induced depression of void swelling in solids.

Material simulations of high-energy radiation environments, such as those existing in outer space¹ and inside nuclear reactors², are usually performed experimentally³ and theoretically⁴ to investigate the behaviour of lattice point defects, i.e. vacancies and interstitials⁵. These primary defects govern not only material microstructural changes, such as the formation of dislocation loops and voids⁶-⁹, but also material mechanical property changes, for example embrittlement¹⁰, creep¹¹ and hardening¹². Although simulation studies have been performed for decades, reports of microstructural changes caused by the simultaneous presence of SVs and FPs are limited, except under ion-electron dual-beam irradiation¹³. Here we show the striking effects of simultaneous irradiation with a laser-electron (photon-electron) dual-beam on void formation in an in situ experiment using a laser-HVEM¹⁴,¹⁵. The laser-HVEM simultaneously introduces SVs and FPs into crystalline solids, as a result of the high intensity photon irradiation from the laser source and the electron irradiation from the HVEM¹⁶, respectively.

Void formation and swelling introduced by irradiation have attracted considerable attention since their discovery in SUS316 steels in 1960¹⁷-¹⁹. SVs and interstitials simultaneously form in materials exposed to outer space or the inside of nuclear reactors through irradiations (neutron, heavy ion, γ-ray and electron), but experimental simulations of this condition have not been accomplished because of the technical limitations associated with nano-scale in situ observations. The use of in situ observation to realise this simulation is essential to understanding void formation and swelling in the outer space or the inside of nuclear reactors.

Since the first report in 1960⁰, laser technology has been widely applied in various fields, such as surgery²¹, radar²², welding²³, material surface modification²⁴ and semiconductor material science²⁵. As a result of their affordability, laser heads can be combined with other equipment to investigate new methods for exploring unknown scientific fields, such as the newly developed laser-HVEM used for materials studies²⁶. The primary advantages of laser-HVEM can be summarised as follows. First, in situ observations of the evolution of material microstructures under laser irradiation are possible²⁶. This method can directly record the evolution procedure and may also reveal the driving mechanism. Second, the evolution of defect structures under laser irradiation provides new measurements for defect migration energy²⁶ as a function of annealing temperature and time. These measurements have led to new lattice defect studies after little activity in the field for more than 30 years. Third, by precisely controlling the laser intensity and the electron dose rate, a laser-HVEM makes it possible to introduce randomly distributed SVs and FPs simultaneously into a crystalline solid. This approach is different from electron irradiation and cascade irradiation (neutron or heavy ions). During electron irradiation, only FPs are introduced, and no cascade damage results²⁶. In the cascade irradiation process, the primary results of the cascade damage are...
high concentrations of vacancies surrounded by interstitials\textsuperscript{27}. Using a laser-HVEM, \textit{in situ} observations of lattice defects under a photon-electron dual-beam irradiation process, which have never been reported to date, are possible.

SUS316L austenitic stainless steel is generally used for core- internals in boiling water reactors; in this study, SUS316L austenite steel was experimentally investigated to determine the effect of laser-electron dual-beam irradiation on void formation and swelling. The results were compared to those for steel subjected to laser-electron sequential irradiation and electron irradiation. Defect rate equations from various irradiation processes were proposed for a theoretical analysis of lattice point defect behaviours. We anticipate not only that the results will provide insight into the mechanism underlying the dual-beam radiation-induced depression of void swelling in solids but also that laser-HVEM will be employed for broader materials research in the future, such as simulation of the environment of outer space, laser-assistant welding and laser fabrication.

**Results**

Figure 1(a), (b), (c), (d) and (e) shows that void formation differs significantly under various irradiation conditions as mentioned in Table 1. For example, under electron (\(e^-\)) irradiation (Fig. 1(a)) and laser-electron (L\(\rightarrow\)\(e^-\)) sequential irradiation (Fig. 1(c) and (d)), a large number of voids are clearly observed; only a few very small voids form under laser irradiation, as the magnified image shows in Fig. 1(b); Fig. 1(e) shows that under laser-electron simultaneous dual-beam (L\(+e^-\)) irradiation, void size seems smaller than that of \(e^-\) irradiation. Individual diagrams of the irradiation procedure are provided under each image to better illustrate the process. Statistical results for the void mean size, number density and swelling are shown in Fig. 2(a), (b) and (c), respectively.

The voids formed under \(e^-\) irradiation alone were found to have a mean size of 21.8 nm and a number density of \(1.4 \times 10^{20} \text{ m}^{-3}\). Under laser irradiation, voids still formed but were found to have the smallest mean size (4.4 nm) and lowest number density (\(0.7 \times 10^{20} \text{ m}^{-3}\)) among the different irradiation methods investigated. Under L\(\rightarrow\)\(e^-\) irradiation, the voids were found to be much larger than the voids formed using laser irradiation alone, and the void number densities were much higher than those observed for \(e^-\) irradiation alone. This result indicates that pre-laser irradiation enhances void nucleation, and subsequent electron irradiation enhances void growth under the present experimental conditions. The mean void size for L\(+e^-\) irradiation is smaller than that of \(e^-\) irradiation and L\(\rightarrow\)\(e^-\) irradiation. However, the void number density is still much higher than that of \(e^-\) irradiation.

Statistical results of void swelling representing the comprehensive results of void mean size and number density are shown in Fig. 2(c). It is clear that under L\(\rightarrow\)\(e^-\) irradiation, the void swelling is much greater than that with \(e^-\) irradiation, suggesting that L\(\rightarrow\)\(e^-\) irradiation enhances void swelling. In the case of L\(+e^-\) irradiation, void swelling was only 15\% and 10\%, respectively, of that observed for \(e^-\) irradiation and L\(\rightarrow\)\(e^-\) irradiation. This illustrates the striking phenomena of (L\(+e^-\)) \(< \ e^- \ < \ \text{L}\rightarrow\text{e}^-\) regarding void swelling.

**Discussion**

A theoretical explanation of defect behaviour during \(e^-\) irradiation has been proposed by Kiritani \textit{et al.}\textsuperscript{28} and by Sizmann\textsuperscript{29}. FPs are introduced during \(e^-\) irradiation, and equations for the concentra-
tion variation of the vacancies $C_v$ and the interstitials $C_i$ are given as follows:

$$\frac{dC_v}{dt} = K - R_{IV}(D_I + D_V)C_IC_V - S_VC_V,$$  (1)

$$\frac{dC_i}{dt} = K - R_{IV}(D_I + D_V)C_IC_V - S_IC_I,$$  (2)

where $K$ is the point defect production rate under $\epsilon^-$ irradiation, $R_{IV}$ is the capture site number of the recombination of the interstitials and vacancies, $S_v$ and $S_i$ are the sink strengths of the vacancies and interstitials, respectively, and $D_V$ and $S_I$ are the diffusivities of the vacancies and interstitials, respectively. The time dependences of $C_v$ and $C_i$ are shown in Fig. 3(a). The dark line represents $\epsilon^-$ irradiation with $5 \times 10^{14}$ m$^{-2}$ which is a general magnitude under $\epsilon^-$ irradiation. Because FPAs are introduced by $\epsilon^-$ irradiation, $C_v$ and $C_i$ continuously increase until the vacancies and interstitials begin to recombine at $t_1$. During the $t_1 - t_2$ period, $C_i$ begins to decrease because of the sink, which contributes to the annihilation of the interstitials, whereas $C_v$ increases because of the lower mobility of the vacancies. A steady-state region occurs at $t_3$: at this point, the defects have equivalent annihilation rates because the sink starts to contribute to vacancy annihilation.

For laser irradiation, after introduced at the surface of the specimen, all SVs diffuse to the interior and distribute homogeneously inside the specimen$^{16}$. Thus, $C_v$ is rewritten as:

$$\frac{dC_v}{dt} = L - S_v D_v C_v,$$  (3)

where $L$ is the SV production rate under laser irradiation. The recombination term is ignored because no interstitials exist. The time dependence of $C_v$ is shown in Fig. 3(b). Even after a subsequent thermal annealing period of 90 min, SVs prefer to form vacancy-type loops$^{16}$. The SVs appear to be “frozen” and do not contribute to void growth; but might actually nucleate the invisible void nuclei and vacancy-type clusters.

In the case of $L \rightarrow \epsilon^-$ irradiation, during pre-laser irradiation, $C_v$ can be given by eq. (3) and is illustrated in Fig. 3(c) by a red dot line. During subsequent electron irradiation, $C_v$ and $C_i$ can be expressed as:

$$\frac{dC_v}{dt} = K - R_{IV}(D_I + D_V)C_IC_V - S_VC_V,$$  (4)

$$\frac{dC_i}{dt} = K - R_{IV}(D_I + D_V)C_IC_V - S_IC_I,$$  (5)

the time dependences of $C_v$ and $C_i$ are shown in Fig. 3(c). $t'$ is the time from 600 s which is the starting point of subsequent electron irradiation. At the initial stage of subsequent electron irradiation, interstitials firstly annihilate to SVs introduced by pre-laser irradiation, once an interstitial leaves its position from a FP to recombine with a SV, the remaining vacancy becomes a new SV. After

Table 1 | Details of the five experimental irradiation procedures

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<tr>
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<th>Experimental Procedure</th>
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<tr>
<td>Irr. 1</td>
<td>Electron irradiation at 723 K for 20 min. ($\epsilon^-$)</td>
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<tr>
<td>Irr. 2</td>
<td>Laser irradiation at 723 K for 10 min (1200 pulses) followed by thermal annealing at 723 K for 90 min. (L)</td>
</tr>
<tr>
<td>Irr. 3</td>
<td>Laser irradiation at R.T. for 10 min (1200 pulses) followed by electron irradiation at 723 K for 20 min. (L R.T. $\rightarrow$ $\epsilon^-$)</td>
</tr>
<tr>
<td>Irr. 4</td>
<td>Laser irradiation at 723 K for 10 min (1200 pulses) followed by electron irradiation at 723 K for 20 min. (L [723 K] $\rightarrow$ $\epsilon^-$)</td>
</tr>
<tr>
<td>Irr. 5</td>
<td>Laser-electron simultaneous dual-beam irradiation at 723 K for 20 min. (L+$\epsilon^-$)</td>
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Figure 2 | Statistical results for the void size (a), number density (b) and swelling (c) of the specimen under five different irradiation conditions. The error bars represent the standard deviation.
the annihilation, interstitials enter a very quick build-up stage; accordingly, \( C_v \) also increases. Because the increment is relative much smaller, the build-up stage of \( C_v \) is marked as the dark dot A in the line. Compared with the \( e^+ \) irradiation (without pre-laser irradiation, as shown in Fig. 3(c) by the dark dot line), it is obvious that before the steady-state \( C_v \) of \( L+e^+ \) irradiation is higher than that of \( e^+ \) irradiation. Consequently, void number density of \( L+e^+ \) irradiation is higher than that of \( e^+ \) irradiation.

By eqs. (4) and (5), \( L+e^+ \) irradiation finally reaches to the same steady state with \( e^+ \) irradiation; different from the \( e^+ \) irradiation, during \( L+e^+ \) irradiation \( C_v \) does not increase but decrease after the recombination stage by the SVs’ diffusion to sinks because of the high \( C_v \). Voids grow during the entire procedure and therefore are much larger than those observed for \( e^+ \) irradiation. From this finding, it becomes obvious that subsequent electron irradiation enhances void growth, which begins as the electron irradiation perturbs the “frozen” SVs. This mechanism can be explained as follows. During pre-laser irradiation, SVs form stable invisible vacancy-type clusters and void nuclei; the subsequent electron irradiation enhances the SVs migration, thus the “frozen” SVs become movable and contributive to void growth.

One experimental difference between the two types of \( L+e^+ \) irradiation (denoted as Irr. 3 and Irr. 4 in Table 1) is the temperature of the laser used for irradiation. SVs are stable at R.T. but are mobile during the heating procedure from R.T. to 723 K. As a result, new voids nucleate, and these differ from those that become nucleated during pre-laser irradiation. This causes the void number density of Irr. 3 to be higher than that of Irr. 4. Both subsequent electron irradiation processes were carried out at 723 K for 20 min, with the same damage rate being observed for each. Stated another way, the effect of subsequent electron irradiation on perturbing the “frozen” vacancies for void growth is the same. If it is assumed that the growth of void nuclei is homogeneous, the void mean size of Irr. 3 should be smaller than that of Irr. 4 due to the higher void number density of Irr. 3.

In the case of dual-beam irradiation, \( C_v \) and \( C_i \) are given as:

\[
\frac{dC_v}{dt} = (L + K) - R_{SV}(D_t + D_V)C_v - S_vD_vC_v, \quad (6)
\]

\[
\frac{dC_i}{dt} = K - R_{SV}(D_t + D_V)C_v - S_I D_vC_i, \quad (7)
\]

when \( L = 0 \), eqs. (6) and (7) represent the same concentration variations under \( e^+ \) irradiation. The time dependences of \( C_v \) and \( C_i \) during \( L+e^+ \) irradiation are also shown in Fig. 3(a) for the convenient comparison with those during \( e^+ \) irradiation. After laser irradiation the magnitude of sink strength was determined in the order of \( 10^{11} - 10^{12} \) m\(^{-2}\) which is lower than the general value of \( e^+ \) irradiation\(^{16} \), thus lines in the colour of red and green represent dual-beam irradiation with \( S = 2 \times 10^{11} \) m\(^{-2}\) and increasing point production rate under laser irradiation (L), respectively. \( C_v \) increases with \( L \). Dual-beam irradiation has the similar stages with those of \( e^+ \) irradiation. The recombination time of \( L+e^+ \) irradiation is longer than that of \( e^+ \) irradiation, which indicates SV and interstitial recombination is enhanced. Unlike \( e^+ \) irradiation and \( L+e^+ \) irradiation, SVs and FPs are introduced simultaneously during \( L+e^+ \) irradiation. In the interior of the specimen, the interstitials and SVs co-exist nearby, as a result, their recombination is the dominant behaviour during \( L+e^+ \) irradiation. Due to the higher \( C_v \), void number density of

![Figure 3](https://www.nature.com/scientificreports/srep01201/images/fig3.png)

**Figure 3** | Calculated time dependences of \( C_v \) and \( C_i \) under different irradiation conditions. (a) Electron irradiation and laser-electron dual-beam irradiation for 20 min. (b) Laser irradiation for 10 min. The SV production rate increases with laser intensity, as the dot line shows. (c) Laser irradiation for 10 min followed by electron irradiation.
L$^+$$^-$ irradiation is higher than that of e$^-$ irradiation. But the void growth is delayed because of the delayed steady-state of L$^+$$^-$ irradiation; therefore, the void size is much smaller than that of e$^-$ irradiation and L$^+$e$^-$ irradiation, which causes the void swelling to decrease greatly. The details of all these calculations are given in the SI. Figure 4 (a), (b), (c) and (d) presents the models of how defect behaviour varies with the irradiation conditions.

In conclusion, SVs introduced by laser irradiation enhance void nucleation but contribute less to void growth. Subsequent electron irradiation perturbs the SVs, causing them to contribute to void growth and to thus increase swelling significantly. SV and interstitial recombination is greatly enhanced by L$^+$e$^-$ irradiation and acts as the dominant governing mechanism of the defect behaviour. Consequently, the degree of swelling due to L$^+$$^+$e$^-$ irradiation is less than that due to e$^-$ irradiation or L$^+$e$^-$ irradiation. Therefore, L$^+$e$^-$ irradiation is shown to depress void swelling substantially in crystalline solids.

**Methods**

The chemical composition of the SUS316L austenite stainless steel used in this study is shown in Supplementary Table 1. The irradiated area of specimen had a thickness of 300–550 nm.

Five different irradiation processes were performed using the laser-HVEM equipment to investigate the effect of laser irradiation on defect behaviour and void formation. The experimental procedures are given in Table 1. The central wavelength of the Nd: YAG laser was 532 nm with a repetition rate of 2 Hz, and pulse duration was 5–6 ns. The average energy density of the laser beam was measured at 24 mJ/cm$^2$. The laser beam energy used in this study to introduce defects inside the specimen was significantly lower than that of the laser-ion dual-beam irradiation process used for controlling nanoparticle precipitates at the surface of semiconductors.

Laser and electron irradiations were carried out at 723 K under a pressure of 10 mN/m. During the laser irradiation, the electron beam intensity for transmission electron microscopy (TEM) observation was kept as low as possible to avoid any effects of this additional irradiation on the specimen. After irradiation, the voids were analysed using a JEM-2000ES TEM.

![Diagram of the defect behaviors under different irradiation conditions.](image-url)

**Figure 4** | Diagrams of the defect behaviors under different irradiation conditions. (a) Electron irradiation. FPs are introduced by electron irradiation; interstitials easily migrate to sinks, while vacancies gradually gather to form voids. (b) Laser irradiation. SVs are introduced at the surface of the specimen by laser irradiation, and subsequently diffuse to the interior and form into void nuclei, of which only a small number grow during the thermal annealing process. (c) Laser-electron sequential irradiations. Effect of pre-laser irradiation is the same as in Fig. 4(b), but subsequent electron irradiation perturbs the SVs, which leads to void nucleation. (d) Laser-electron dual-beam irradiation. SVs and FPs recombine because they are introduced simultaneously by laser irradiation and electron irradiation, respectively. Although void nucleation is abundant, growth is inhibited, and the void mean size is consequently smaller than that observed under any other conditions.


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Author contribution
Z.B.Y. designed and performed the experiment, collected and analysed data and wrote the paper; S.W. designed the study, analysed data and assisted with writing the paper; T.K. was involved in the analysis. All authors discussed the results.

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