

Atomistic investigation of cavitation and ablation in tantalum foils under irradiation with x-rays approaching 5 keV

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The rapid irradiation and heating of matter can lead to material removal via a process known as ablation. Whilst previous investigations have focused on ablation with optical and soft x-ray pulses, the process is not well understood for the high energy x-rays delivered at current XFEL facilities. In this article, we use hybrid two-temperature model molecular dynamics (TTM-MD) simulations to determine the damage threshold and dynamics for tantalum foils under irradiation with x-rays in the range 1-5 keV. We report that damage occurs for foils with thickness ≥ 300 nm when heated to around 1.25 eV / atom. This damage results from the combined processes of melting and cavitation, finally resulting in the removal of material layers. The predictions of this study, in terms of the cavitation threshold and underlying dynamics, could guide interpretation of experiments as well as applications including development of beamline optics for free-electron lasers. We report consistency between cavitation and ablation behaviour in isochoric heating experiments and spall processes in hydrodynamic compression and release experiments, confirming the primary modes of damage are mechanical in nature for the x-ray energies investigated.

I. INTRODUCTION

Irradiation of materials by short pulses of energetic radiation leads to the removal of layers of matter, and localized destruction, via a process known as ablation. This is a process of interest to industry due to its capability to imprint micro-structures on surfaces¹⁻⁴, as well as the potential to improve beam-line optics through a deeper understanding of the mechanisms by which damage occurs under intense irradiation. The minimum fluence at which damage occurs in a material is the ablation threshold, the measurement of which can provide information on material properties as well as on the parameters of the incident radiation. For example, extensive work has been conducted in Refs. 5–10 to the use of the ablation profile imprinted on materials by XUV and hard x-ray pulses to diagnose the spatial characteristics and transverse intensity profile of the incident beam. Numerous experimental studies have reported on the ablation threshold for a range of metals under irradiation spanning a wide range of wavelength and pulse duration^{1,11–14}. Despite these efforts there remain gaps in the theoretical understanding of the ablation process, particularly under hard x-ray irradiation.

The dynamics of the x-ray matter interaction are strongly dependent on the properties of the material and the characteristics of the incident irradiation. There are, however, general features of the irradiation and subsequent ablation processes, which will be briefly outlined. The incident x-ray photons are scattered or absorbed, depending on the x-ray energy and the atomic number of

the material. For transition metals such as tantalum, interacting with ≈ 10 keV photons, absorption is the dominant process. The x-rays interact with the inner shell electrons, resulting in liberation of photo-electrons, fluorescence or the emission of Auger electrons (which are more important in low- Z elements). These processes result in rapid redistribution of the electron energy, and equilibration of the free electron gas over a time scale of ≈ 100 femtoseconds (fs). In the case of very short pulses of intense radiation (for example, the ≈ 100 fs pulses deliverable by XFELs), the electrons reach extremely high energy densities following irradiation. The transfer of heat from the electrons to the lattice occurs over a much longer time scale, of the order 10s of picoseconds (ps). This rapid heating of the lattice occurs faster than the time scale associated with thermal expansion, which is determined by the speed of sound (typically in the range 10-100 ps). Heating therefore occurs under isochoric conditions, resulting in the formation of a large thermal pressure within the material. This isochoric heating can also occur in materials irradiated with optical and UV radiation^{15–17}. Specifically during hard x-ray irradiation, there is the complication that the emitted photo-electrons change the internal charge distribution. This can lead to Coulomb forces playing an important role in the ablation process, and the resulting damage dynamics are not well understood¹⁶.

Following the isochoric heating stage, the thermal pressure within the sample is released over the time scale of thermal expansion. The dynamics of the pressure release

depend on the uniformity of the heating. Existing studies have reported on the conditions under which metals undergo ablation following irradiation with optical and soft x-ray pulses^{1,18–24}. Due to the short penetration depth of such radiation, only a small volume near the surface of the metal is heated. The resulting localized thermal pressure is released by launching a leading (compressive) wave into the bulk of the material, accompanied by a trailing (rarefaction) wave propagating from the front surface as it expands into vacuum. The superimposed wave has a bipolar spatial profile and propagates towards the rear surface. This wave is subsequently reflected, with the leading component transitioning into a tensile one and overlapping with the incoming trailing wave to form a region of tension within the metal^{19,25}. For a sufficiently high degree of heating (and therefore release of a large thermal pressure), the tensile stress may exceed the tensile strength of the material. Note that this situation, where the release of pressure drives a bipolar wave into the material, is analogous to the seminal work by Thomsen²⁶, who derived the equations governing the propagation of waves in a weakly anisotropic medium.

In contrast, if the entire metal is uniformly heated (and there is no pressure gradient), the pressure waves do not exhibit the bipolar spatial profile described previously. Instead, rarefaction waves are released from the upstream and downstream free surfaces simultaneously as they expand into vacuum. This uniform heating can occur even when the initially heated volume is small if the material has a large thermal conductivity, or if heat is carried deep into the bulk by highly energetic electrons.

Thus, for materials subjected to hard x-ray irradiation, the uniformity of the heating falls between the two extremes described above. From the experimental perspective, the thickness of material is often chosen to be approximately equal to the absorption depth of the incident radiation, i.e. the distance over which the electron temperature decreases to $1/e$ of the surface value. This condition is a compromise between having a sufficiently thick sample that there is a strong scattering signal, without being too thick that both the incident and scattered x-rays are absorbed, resulting in a decrease in signal at the sample rear. This condition results in a pressure and temperature gradient across a greater extent of the material compared to the case for weakly penetrating radiation. The propagation of the pressure waves from the free surfaces is further complicated by the fact that the temperature gradient in the sample means different regions have different tensile strength limits. This can lead to localized ablation²⁷, which does not necessarily occur at the centre of the material as it does for uniform heating.

The formation and decomposition of a stress confined state under intense heating via pulsed radiation exposes the complexity of the ablation dynamics. Whilst experiments are only partly sensitive to these dynamics, hybrid two-temperature model molecular dynamics (TTM-MD) simulations can explore them in detail^{28,29}. In contrast to ‘standard’ MD, the TTM-MD approach enables en-

ergy to be coupled to an electron subsystem and slowly transferred to the lattice. The unique advantage offered by this approach is that, unlike finite-difference and hydrodynamic simulations, it is possible to track individual atomic trajectories throughout the interaction. This is particularly important for high energy ablation processes. The applicability of the TTM-MD approach has been demonstrated in recent computational studies, where the predictions of the ablation threshold and crater depth show agreement with experimental measurements^{19,21}.

A limitation of the aforementioned studies is that they are restricted to the regime of weakly penetrating radiation^{1,18–24}. We aim to extend the simulation approach to incorporate higher energy x-rays, in the range 1–5 keV, where the ablation process is poorly understood. This energy is within the operating range of current XFEL facilities and provides a means of interpreting recent experimental results. Whilst previous studies have investigated the pressure-induced phase transitions³⁰ and tensile strength of tantalum using simulations of shock compression and release^{27,31–33}, here we specifically simulate the isochoric heating and subsequent pressure release which leads to material failure.

This article presents numerical simulation results which explore the cavitation threshold and subsequent ablation dynamics for tantalum foils under intense x-ray irradiation (up to ≈ 5 keV). In Section II, an overview is provided of the different mechanisms by which ablation occurs under short pulse irradiation. This is followed in Section III by a discussion of the TTM-MD approach implemented in the open-source molecular dynamics code, LAMMPS³⁴, employed in this study. The key results, namely the criteria for destruction of foils of different thickness values, are presented in Section IV. Finally, the results are summarised in Section V along with a discussion of their wider impact and relevance to XFEL experiments.

II. UNDERPINNING THEORY

II.A. Ablation mechanisms under femtosecond pulse irradiation

There are numerous mechanisms which can result in the destruction of material under pulsed radiation. The exact mechanism which dominates an interaction depends on the duration of the radiation pulse, as well as material properties including the speed of sound, thermal conductivity and in the case of metals, the rate of energy transfer from the electron subsystem to the lattice. A complete understanding of the process of material removal (ablation) is further complicated by the fact that the mechanisms can feed into each other, operating in different regions of the material on the same time scale³⁵. In this section, a brief overview is provided of the ablation mechanisms present in materials under ultra-short pulse irradiation.

As discussed in the introduction, femtosecond pulse ir-

radiation results in the formation and decomposition of a stress confined state. Subsequent pressure release creates tension, which can lead to failure of the material if the tensile strength is exceeded. Otherwise, the pressure waves continue to reverberate within the sample and can, for example, be used to generate off-Hugoniot states in layered samples³⁶. If the material remains solid up to the point of failure, this process is known as spallation. The phase diagram for tantalum is plotted in Fig. 1, where the black curve labelled (1) indicates a typical thermodynamic trajectory associated with spallation as described in Refs. 37 and 38. The process begins with isochoric heating, indicated by the vertical section of the trajectory. This heating is insufficient for melting, and the trajectory is subsequently constrained to the solid region of the phase diagram.

The high levels of lattice heating achievable under femtosecond irradiation mean that the material is often heated above its melting point. Analogously to the process of spallation in solids, liquid metals can undergo failure when they are subjected to tensile stress which exceeds their tensile strength. This process is known as cavitation. It resembles boiling, in that it proceeds through the formation of voids or cavities in the material, but at temperatures below the boiling point^{25,39}. The process of cavitation is possible due to tensile stresses within the liquid, which drive the void formation.

The thermodynamic trajectory for cavitation is plotted in Fig. 1 and denoted by the curve labelled (2). Following the initial isochoric heating stage, the sample undergoes isentropic cooling and crosses into the vapor dome between the triple and critical points. The material then exists as a metastable liquid, which will eventually transition into a mixed liquid-vapor phase if an energy barrier is overcome^{40,41}. This barrier is that the pressure in the vapor phase must exceed the pressure in the liquid plus the pressure associated with the surface tension across the interface separating the phases⁴². This is written in Eqn. (1) below.

$$P_v > P_l + \frac{2\sigma}{r} \quad (1)$$

In the above equation, σ is the surface tension measured in units of Jm^{-2} and r is the radius of the void. Equation (1) suggests there is a critical radius where the probability of the forwards and backwards phase transitions are equally likely⁴¹. This critical radius has been evaluated for Cu to be 3-4 Å in Ref. 43 and 7.3 Å in Ref. 41, using a combination of classical nucleation theory and MD simulations. We do the same for tantalum in the Appendix, yielding a value of 4.2 Å. Whilst this radius denotes the size at which the void is likely to continue growing, there is still a probability that a critically sized void will collapse at later times⁴¹. This is especially relevant in a system where reverberating pressure waves can disrupt the equality in Eqn. (1), which is the case for the foils in this study.

The process of ‘normal’ boiling is not possible on the timescale of a femtosecond pulse, as noted in Ref. 44.

Instead, the process of ‘phase explosion’ (or ‘explosive boiling’) becomes dominant. In this case, a layer of material near the irradiated surface is heated to its stability limit, typically understood to occur at $0.9T_c$ where T_c is the temperature at the critical point³⁵. The material exists as a super-heated liquid, i.e. it remains in the liquid state but is heated above the boiling point, and phase explosion is the rapid phase transition from this super-heated liquid to a two-state mixture of liquid and vapor³⁵. There are a number of signatures which characterize the phase explosion process, including; a sharp increase in the yield of ablated atoms⁴⁵, a change in the ablated plume from distinct liquid layers to a mixture of vapor-phase atoms and clusters^{35,37,38}, and a high nucleation rate as the trajectory approaches the spinodal line (the stability limit)³⁷.

In this article we follow the thermodynamic trajectory of sections of the foil (which are representative of the average bulk material) through phase space to diagnose the ablation mechanism. This approach is outlined in Refs. 37, 38, 46, and 47. We note that Refs. 37 and 38 are limited by the use of 2D simulation geometry and make incorrect predictions about the conditions for phase explosion. Here we use fully 3D simulations and limit our discussion to lower energy mechanisms (spallation and cavitation), which can be reasonably inferred by investigating whether or not the trajectory crosses the melt curve and subsequently progresses into the vapor-dome (measured for the chosen MD potential). We do not typically consider phase explosion given that the level of heating is well below the critical temperature, even close to the upstream free surface where the temperatures are highest.

III. METHODOLOGY

III.A. Computational model

In this investigation we employ hybrid two-temperature model molecular dynamics (TTM-MD) simulations, performed with the open-source code LAMMPS³⁴. The development of the TTM module for LAMMPS is detailed in Refs. 53 and 54 and the underlying algorithm is described in Refs. 28 and 29. In these TTM-MD simulations, the atomic lattice is overlaid with a grid of ‘electron’ cells. The radiation energy is deposited into the grid, with the temporal evolution of the grid temperature determined by solving the heat conduction equation, shown below, using a finite difference scheme.

$$c_e \rho_e \frac{\partial T_e}{\partial t} = \nabla \cdot (\kappa_e \nabla T_e) - G(T_e - T_a) + \frac{I_0}{\ell_s} \exp\left(-\frac{x}{\ell_s}\right) \quad (2)$$

In the above equation c_e is the electron heat capacity and κ_e is the electron thermal conductivity. It is important to note that, in LAMMPS, it is possible to incorporate the dependence of both c_e and κ_e on the electron

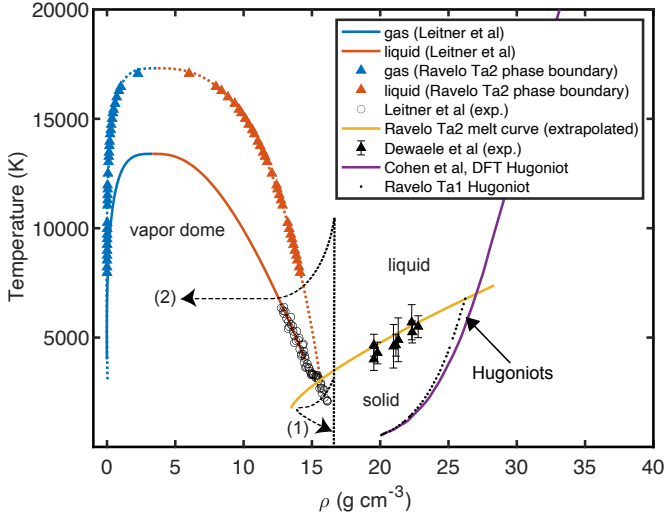


FIG. 1 Tantalum phase diagram. The solid blue and orange curves indicate the vapor dome boundaries obtained from Ref. 48. These boundaries are extrapolated from the experimental data points marked by open circles at the liquid end of the phase boundary. The blue and orange triangles mark the vapor dome boundaries (i.e. the binodal line) obtained from mixed phase simulations (described in the Appendix) using the Ravelo Ta2 potential from Ref. 31. The dotted blue and orange curves are fitted to the mixed phase simulation data points, using the same fitting equation as Ref. 48. The yellow curve is the melt curve for the Ravelo Ta2 potential used in this study, with black triangles indicating the experimental data points from Ref. 49 used in the fitting of this EAM potential. The melt curve is extrapolated to negative pressure values using the fitting equation presented in Ref. 50, and converted to density-temperature space using the EoS spreadsheet in Ref. 51. The Hugoniot (purple curve) is obtained from DFT calculations in Ref. 52, whilst the dotted black curve is the Hugoniot calculated using the Ravelo Ta1 potential in Ref. 31. The vertical dashed line is the ambient density isochore for tantalum, with branches (1) and (2) representing thermodynamic pathways associated with spallation and cavitation, adapted from Ref. 37.

temperature. The electron temperature and density are denoted T_e and ρ_e respectively, whilst T_a is the atomic (lattice) temperature. The rate of heat transfer from the electrons to the atoms is determined by the electron-phonon coupling constant, G , which has been calculated for a range of materials and as a function of electron temperature in Refs. 55–57, and measured specifically for tantalum in Ref. 58. The final term on the right of Eqn. (2) is the radiation source term²³. The absorbed intensity is denoted I_0 and the term ℓ_s is the absorption depth. The energy is only deposited into electron cells with non-zero temperature, i.e. there is no energy exchange with the vacuum regions. As the foil expands into vacuum, the electron cells in the regions which were previously unoccupied by atoms, obtain a non-zero temperature. This subsequently enables heat exchange with neighbouring (non-vacuum) cells. The values of all the above parameters used in this study are detailed in the next section.

The electron cells act as finite heat reservoirs for the atoms located in the same volume. Heat is passed from

the electrons to the atoms via a Langevin thermostat. The equation of motion for a given atom (with mass, m_a , and velocity, v_a) is given in Eqn. (3) below.

$$m_a \frac{\partial v_a}{\partial t} = F_a(t) + \tilde{F}(t) \quad (3)$$

The first term on the right is the force exerted on the atom due to the inter-atomic potential, whilst the second term is a force associated with the Langevin thermostat. The electron cell acts as a finite heat reservoir, giving ‘kicks’ to the trajectory of atoms located within the same volume, with an amplitude proportional to the temperature of the local electron cell. This interaction simulates the electron-phonon coupling which is responsible for energy transfer from the electron subsystem to the atomic lattice.

Following the method prescribed in Ref. 53 the TTM module runs until the temperatures of the electron subsystem and the lattice reach equilibrium (when the average electron and lattice temperatures in the foil are within 100 K of each other), at a time denoted τ_{eq} . This time depends on the energy deposited into the electron subsystem, with τ_{eq} increasing with the deposited energy. For the range of deposited energy under investigation, τ_{eq} occurs before 30 ps, which is selected as the TTM cut-off time for these simulations. This is demonstrated in Fig. 2 where the average electron and lattice temperatures within a 1000 nm thick foil are plotted, for the case of heating to above the ablation threshold. The dashed blue and solid blue curves indicate the average electron and lattice temperatures within the foil, respectively. The solid horizontal line in Fig. 2 indicates the melting point measured for the embedded atom model (EAM) potential used in this study, at the peak value of hydrostatic pressure for this particular simulation (which is $P=23.4$ GPa). The vertical dashed line corresponds to the time at which the average electron and lattice temperatures reach equilibrium. The average electron temperature decreases exponentially with time, with a characteristic $1/e$ time scale of 8.5 ps. The vertical line then indicates that electron-lattice temperature equilibration occurs within approximately 2.7 e-foldings. It is therefore appropriate to choose a cut-off time for the TTM module equal to 30 ps, or 4 e-foldings. In addition, the orange curve is the rate of lattice heating, which has slowed to approximately zero by 20 ps. These diagnostics confirm that the TTM module can be switched off at 30 ps without affecting the dynamics of the atoms, given that the lattice has reached thermal equilibrium with the electron subsystem by this time. Again it is important to stress that the selected TTM cut-off time of 30 ps is shorter than the typical hydrodynamic relaxation time scale, meaning that heating occurs under isochoric conditions.

Previous studies have utilised TTM-MD simulations to investigate ablation of metals under pulsed radiation. These may be grouped together depending on the penetration depth of the incident radiation and the material under investigation. For example, Refs. 20, 21, 23, and

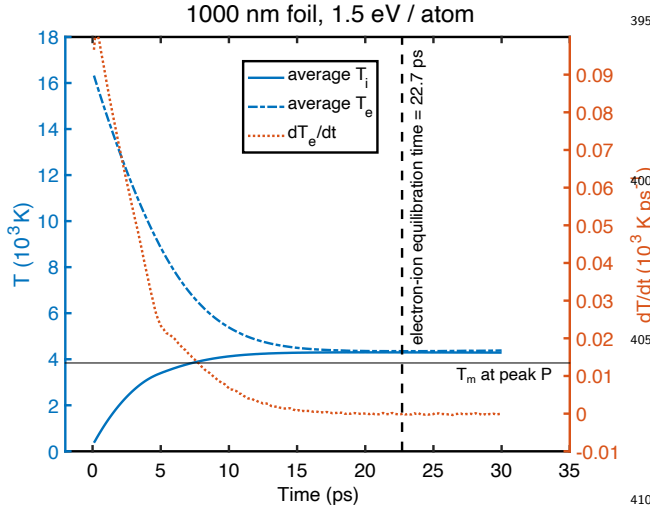


FIG. 2 Electron and lattice temperatures during the two-temperature model stage of the simulation for a 1000 nm thick foil heated to 1.5 eV / atom. The temperature of each species is averaged over the initial dimensions of the foil. The solid blue curve is the average lattice temperature within the foil, whilst the dashed blue curve is the average temperature of the electron subsystem. The orange dotted curve on the right hand axis is the rate of change of the lattice temperature. The black horizontal line is the melting temperature for the EAM potential, at the value of peak pressure within the simulation. These diagnostics indicate that, at a time of 30 ps, the temperatures of the electron subsystem and the lattice have equilibrated, and the rate of lattice heating has slowed to zero.

59 model the ablation of gold irradiated with weakly penetrating (optical or soft x-ray) radiation, whilst Refs. 18 and 24 investigate ablation of thin gold foils in which uniform energy deposition is employed to simulate the role of ballistic electrons. Another commonly studied metal is nickel, for example, in Refs. 22 and 25 nickel ablation is investigated using short pulses of weakly penetrating radiation. The strength of the TTM-MD technique is reflected in the fact that the studies detailed in Refs. 19 and 21 found agreement between the predicted ablation threshold and the experimental measurement in gold. In addition, Ref. 1 reports that the predicted crater depth agrees with experimental measurements for both optical and soft x-ray irradiation of gold foil. However, it is important to note that the aforementioned studies have employed weakly penetrating radiation. A notable exception is Ref. 2, in which the possibility of structuring gold surfaces with high energy x-rays is discussed. In that study the absorption depth was 100 nm, corresponding to approximately 1 keV x-rays propagating in ambient density gold. However, current XFEL facilities are capable of delivering exceptionally bright pulses of hard x-rays with photon energies up to 25 keV, indicating a necessity to extend TTM-MD simulations to higher x-ray energies.

In this study we model the effect of the high energy x-rays by increasing the absorption depth of the incident radiation (ℓ_s in Eqn. (2)). This is an attractive option for investigation given that it requires varying only the absorption depth and the incident intensity (to reach

the desired level of lattice heating), and does not require modification of the source code to implement the high energy radiation. There is however a limit to this approach, in that the equations of the standard TTM do not address some of the complexities of hard x-ray matter interactions. For example, this approach does not explicitly account for non-thermal ballistic electrons within the metal. Here, the term ‘non-thermal’ refers to the fact that the ballistic electrons are not in local thermal equilibrium with the finite heat reservoirs which comprise the electron subsystem. These ballistic electrons are seeded by intense x-ray irradiation and can carry heat much deeper into the metal than the absorption depth would imply. They are predicted to play an important role in studies of noble metals but a much lesser role for transition metals such as tantalum^{60,61}. It is suggested in Refs. 18 and 24 that the effect of the ballistic electrons can be included by modifying the heat source term to give uniform energy deposition across the material, accounting for the deeper penetration of the heat.

Given the limitations detailed above, we restrict the x-ray energies in this study to the range 1-5 keV. These energies are still within the operating range of current XFEL facilities. Additionally, the pressure and temperature regime accessible in the simulations is comparable to the regimes reported in recent experimental and simulation-based studies (Refs. 27 and 39), providing a point of comparison for our results.

As the fluence increases, photo-ionization effects (such as multi-photon and barrier suppression ionization⁶²) also become important, which again are not included in TTM-MD simulations. This restricts the regime of validity to below the plasma formation threshold. However, we have no necessity to model these effects here, given that the mechanisms by which destruction of the foils occur (cavitation and subsequent ablation) are present well below the plasma formation threshold.

III.B. Simulation parameters

The simulations detailed in this paper are for free-standing tantalum foils, with thickness ranging from 150 nm to 1000 nm. In each case, the absorption depth (the depth at which the electron temperature decreases to $1/e$ of the peak value) is set equal to the foil thickness. As discussed in the introduction, this approximates experimental conditions, in which the foil thickness is chosen to be close to one absorption length for the incident radiation. Varying the absorption depth in this way is equivalent to changing the energy of the incident radiation. For the range of absorption depth values in this study, the energy of the x-ray pulse is in the range 1.3 - 4.9 keV, estimated using data from Ref. 63.

The remaining pulse parameters are as follows. The pulse temporal profile is a top-hat function with a pulse duration of 100 fs. The LAMMPS TTM module requires the user to define the absorbed fluence, i.e. the fluence which is incident on the front surface of the foil. The

fluence values in this study are converted to an average atomic temperature in eV / atom. This enables the ablation threshold to be determined in terms of the average lattice temperature as opposed to stating the threshold as a fluence value, which depends on the absorption depth and foil thickness (although the fluence is also reported for convenience in the text). For each foil thickness, four simulations are run, corresponding to lattice heating to 0.95, 1.25, 1.5 and 2 eV / atom. This range captures heating from just below the melting point up to the predicted boiling point for the tantalum EAM potential, and therefore straddles the ablation threshold.

The simulations use the EAM potential developed by Ravelo *et al.* in Ref. 31. EAM potentials are particularly well suited to molecular dynamics simulations of a free-electron gas, since they account for many-body effects whilst maintaining a relatively low computational cost. It is reported that the computational cost of many-body potentials in molecular dynamics simulations scales linearly with the number of atoms⁶⁴. This facilitates simulations with 10s of millions of atoms, which are necessary in the current study, given the absorption depth of the hard x-rays under consideration is approximately one micron.

Laser-induced ablation involves a combination of quantum (laser absorption and electron-phonon coupling) and classical (hydrodynamic release, melting and tensile strength) processes, meaning that it is extremely difficult to locate a potential which accurately captures all of the underlying physics. We tested a range of tantalum EAM potentials in preliminary simulations, however the Ravelo Ta2 potential was determined to perform best for this investigation. Two important considerations are the fact that the Ravelo Ta2 potential gives a smooth variation of the Grüneisen parameter as a function of volumetric compression, even at the high pressures and temperatures accessible in this study, and that the melt curve for this potential is in agreement with experimental data obtained from diamond anvil cell studies. The experimental results against which this potential are validated extend up to 100 GPa, as described in Ref. 49. It is worth noting that the Ravelo Ta2 potential underestimates the Grüneisen parameter by approximately 40% at ambient conditions, which will subsequently have an effect on the thermal expansion coefficient of tantalum³³. However, it is reported in Ref. 33 that the average value of the Grüneisen parameter from the simulations in the pressure range 0-100 GPa is close to the average over the corresponding experimental data. Again, we stress that the robustness of the Ravelo Ta2 potential in the high-pressure regime, and its demonstrable ability to recreate the thermal properties of tantalum, make it a suitable choice for this study.

In TTM-MD simulations it is necessary to specify the form of the electron heat capacity, c_e , and electron thermal conductivity, κ_e . Here, we assume the free-electron gas model, in which the electron heat capacity varies linearly with electron temperature, T_e . This linear ap-

proximation is typically valid up to the Fermi temperature, and indeed the peak electron temperatures reached in this study are well below this limit. It follows that $c_e = \gamma T_e$, where $\gamma = 77.4 \text{ J m}^{-3} \text{ K}^{-2}$. In LAMMPS, the electron thermal conductivity is related to the electron heat capacity through the diffusivity constant, D_e , such that $\kappa_e = D_e c_e$. The diffusivity constant is defined as follows:

$$D_e = \frac{K_0}{\gamma T_a} \quad (4)$$

where $K_0 = 57.6 \text{ W m}^{-1} \text{ K}^{-1}$ is a constant, obtained from Ref. 65 and T_a is the atomic (lattice) temperature. The chosen value of K_0 likely leads to an underestimate of the electron thermal conductivity, as noted in Ref. 66, however more recent calculations in Ref. 67 show that our value is only a factor two lower than the theoretical prediction under equilibrium conditions. It is demonstrated in Ref. 67 that the electron thermal conductivity of tantalum has a non-linear dependence on T_e . Given that LAMMPS relates the thermal conductivity to the heat capacity via the thermal diffusion constant, it is not possible to implement an electron thermal conductivity with a different functional form to that of the electron heat capacity (which Ref. 67 shows is indeed the case), without modification of the source code. We therefore chose the K_0 value described above and leave detailed investigations of the effect of non-linear electron thermal conductivity for future work. The final thermodynamic parameter is the electron-phonon coupling constant, $G = 3.08 \times 10^{17} \text{ W m}^{-3} \text{ K}^{-1}$, obtained from Ref. 58.

The free-standing foils comprise a bcc lattice with 60×60 unit cells in the transverse dimensions, and 453 - 3027 unit cells in the longitudinal (x-ray pulse propagation) direction, depending on the foil thickness. The transverse dimensions of the simulation box greatly exceed the predicted critical void diameter (detailed in the Appendix) and so the growth of super-critical voids is uninhibited by the simulation size. We also note that the ablation threshold, which occurs when a void reaches a size comparable to the transverse dimensions, is reported to have a dependence on the simulation size^{19,68,69}. In contrast, we report that the cavitation threshold (the fluence at which voids appear) is consistent provided the transverse dimensions exceed the critical void diameter, and has been confirmed during convergence testing of our simulation setup.

The simulation effectively models heat conduction only along the x-ray pulse propagation direction. Given that the x-ray pulses employed experimentally typically have a Gaussian spatial profile with full width half maximum diameter $\approx 10 \text{ } \mu\text{m}$, and the transverse dimension of the foil in the simulation is 19.8 nm, there will be negligible lateral heat transport. This justifies the 1D heat conduction model employed in this investigation. The foil sits in the middle third of the simulation box, with vacuum regions on either side, equal to the foil thickness. The simulation box has periodic boundaries in the transverse

dimensions and non-periodic boundaries in the longitudinal direction. The simulation box is overlaid with a grid on which the electron subsystem is defined, where each cell in the grid has a longitudinal extent of 1 nm.

Each simulation is run for sufficiently long that the rarefaction waves propagating from the free surfaces have time to interfere and create a region of peak tension. For the 150, 300 and 500 nm thick foils a run time of 200 picoseconds (ps) is sufficient, however for the 1000 nm simulations the run time is extended to 400 ps. In every case the MD time step is equal to 1 fs, which is sufficient to generate smooth atomic trajectories whilst incurring a minimal computational cost.

Finally we comment on the notation used to report the hydrodynamic variables from the simulations. LAMMPS enables the computation of the components of the stress tensor for each individual atom. The output quantity has units of pressure multiplied by volume, so to obtain a pressure value it is necessary to divide through by the per-atom volume. This quantity is estimated from the simulations by calculating the volume of the Voronoi tessellation around each atom. The spatial profile of each stress tensor component is then computed by binning the per-atom values across the simulation box, summing the values within each bin, and dividing by the sum of the per-atom volumes within the bin. The hydrodynamic pressure, P , is related to the diagonal components of the stress tensor via Eqn. (5), below.

$$P = -\frac{1}{3}(\sigma_{xx} + \sigma_{yy} + \sigma_{zz}) \quad (5)$$

The x-ray pulse propagates along the simulation x -axis, which is also the axis of pressure release. Throughout this article we report the value of hydrostatic pressure when the tensile stress within the foil reaches its peak magnitude. From Eqn. (5) this results in negative values of pressure. We also report the values of the components of the stress tensor noting that in the liquid phase, $\sigma_{xx} \approx \sigma_{yy} \approx \sigma_{zz}$, and that $P \approx -\sigma_{xx}$.

IV. RESULTS

IV.A. 150 nm free-standing foil

The first results presented are from four simulations with a 150 nm thick free-standing foil, heated to 0.95, 1.25, 1.5 and 2 eV / atom, respectively. The 150 nm foil is sufficiently thin that heat is carried across the extent of the target during the electron-lattice equilibration stage. This results in uniform heating, with the electrons and lattice reaching thermal equilibrium by 30 ps. This is demonstrated in Fig. 3, where the temperature profiles are plotted for the case of heating to 2 eV / atom (corresponding to a fluence of 2700 Jm^{-2}). The solid blue and dashed blue curves correspond to the electron and lattice temperatures at the foil front surface. The solid red and dashed red curves correspond to the same quantities on the rear surface. Fig. 3 demonstrates that by 25 ps, the lattice front and rear temperatures have reached

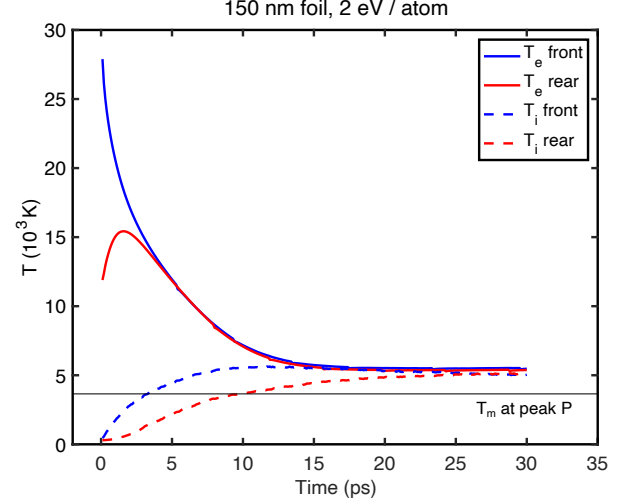


FIG. 3 Temperature profiles for a 150 nm free-standing foil heated to 2 eV / atom. The solid blue and dashed blue curves correspond to the electron and lattice temperatures measured at the foil front surface. The electron and lattice rear temperatures are plotted with solid red and dashed red lines, respectively. The horizontal line is the melting temperature for the EAM potential at the value of peak pressure in the simulation.

equilibrium. This lattice temperature sits well above the melting point of the EAM potential (measured at the value of peak pressure within the sample, indicated by the horizontal black line). As discussed in the previous section, the TTM module is switched off at 30 ps, given that the rate of lattice heating approaches zero.

The peak tensile stress which can be achieved in the 150 nm thick foil is limited by the fact that the condition for heating under stress confinement is only weakly met. As described in Ref. 25, the stress confinement condition is that the radiation pulse duration and the electron-lattice equilibration time must be less than the time scale of thermal expansion, obtained by dividing the foil thickness by the speed of sound. Estimating the speed of sound from the simulation as 3479 ms^{-1} (which is in agreement with the ambient density value of $\approx 3400 \text{ ms}^{-1}$ from Ref. 51), the time scale for thermal expansion is approximately 40 ps, comparable to the electron-lattice equilibration time of 30 ps. Given that the stress confinement condition is only just met, a larger fraction of the foil will have relaxed over the time scale of electron-lattice equilibration, compared to the thicker foils. In this case a relatively low thermal pressure builds within the foil which, upon release, generates tension with a small magnitude. It is not until heating to 2 eV / atom that evidence of cavitation appears in the simulations.

Space-time plots of the density, pressure and lattice temperature evolution are included in Fig. 4, for two different levels of heating. In Fig. 4 (a)-(c) heating occurs to 1.5 eV / atom (at a fluence of 1995 Jm^{-2}), whereas Fig. 4 (d)-(f) corresponds to heating to 2 eV / atom. First, looking at the temperature profile in Fig. 4 (c), it is evident that the front and rear surfaces of the foil have

reached a similar temperature, just exceeding the melting point of the EAM potential. It is inferred that at this point in the interaction the foil is molten, however further evidence is provided in the Appendix, Fig. 15, where the melting of the lattice is corroborated by a broadening of simulated x-ray diffraction peaks. Fig. 4 (c) also shows there is some periodicity in the temperature as a function of time, coincident with oscillations in the pressure and density, due to hydrodynamic reverberations.

The density space-time profile plotted in Fig. 4 (a) shows that the rarefaction waves, travelling from the free-surfaces inwards, begin to overlap at approximately 40 ps. Looking at the space-time profile of the pressure in Fig. 4 (b), it is clear that tension forms within the foil at this time, when the rarefaction waves intersect. The peak tension (i.e the greatest magnitude of σ_{xx}) is reached at the center of the foil at time of 70 ps. However, it is not sufficient at this time to exceed the tensile strength of the material, and the pressure waves continue to reverberate within the foil. The frequency of these oscillations is high compared to the thicker foils, since the pressure waves can traverse the thinner foil more quickly. Comparing Figs. 4 (a) and (b), the pressure oscillations coincide with oscillations in the density; the high density regions align with those of positive hydrodynamic pressure, and vice versa.

As the heating increases to 2 eV / atom, the peak thermal pressure within the foil increases. The release of this pressure results in tension which is sufficient in magnitude to lead to cavitation. The value of hydrodynamic pressure at the time of maximum tension (σ_{xx} reaches its peak value at time $t=62.5$ ps) is $P=-7.63$ GPa. Since the foil is molten at this time, the components of the stress tensor have similar values of $\sigma_{xx}=7.78$ GPa, $\sigma_{yy}=7.59$ GPa and $\sigma_{zz}=7.53$ GPa. Interestingly, we observe anisotropy in the pressure tensor components under weaker heating to 0.95 eV / atom, where the lattice temperature sits below the EAM melting point. In that case, the values of σ_{xx} , σ_{yy} and σ_{zz} are 4.79, 3.32 and 3.43 GPa, respectively.

Strong evidence of cavitation is provided in Fig. 4 (d), where a large void appears near the center of the foil at time of 70 ps, and remains until around 190 ps. The location of this void coincides with the peak tensile stress, further hinting that cavitation is the mechanism underlying failure of the foil. The general trend for the 150 nm foils is that the magnitude of tensile stress increases with the level of heating. The pressure at the time of maximum tension varies from -3.85 GPa to -7.63 GPa as the heating increases from 0.95 to 2 eV / atom.

There is also evidence of additional, smaller voids forming around the largest void. Again, these are seeded in the relatively wide region of tensile stress around the center of the foil. These ‘satellite’ voids which are visible in Fig. 4 (d) collapse at later times and do not lead to ejection of material layers. The sections of material which are separated by these voids subsequently act as independent foils, since void formation results in additional

free surfaces.

Finally, we comment in more detail on the dynamics underpinning the failure of the foil. The density space-time plot in Fig. 4 (d), and further visualization of the atomic positions, demonstrate that all of the voids collapse before the end of the simulation. Despite having a radius greater than the predicted critical value, the reverberating pressure waves likely trigger the void collapse and we do not see ablation, which arises when the voids grow to a size comparable to the transverse dimensions of the simulation box. We conclude that the cavitation threshold for the 150 nm foil occurs at 2 eV / atom, but the voids which form do not lead to splitting of the foil.

IV.B. 300 nm free-standing foil

In contrast to the 150 nm thick foil, the 300 nm foil is sufficiently thick that thermal conduction cannot equalize the lattice temperatures at the front and rear of the foils. This asymmetry in the heating leads to a temperature and stress gradient through the foil. The increased absorption depth in this case also means that a larger volume of the foil remains under stress confinement at the electron-lattice equilibration time. This results in the release of a larger thermal pressure, inducing a state of tension with greater magnitude relative to the 150 nm thick foil.

Space-time plots of the density, pressure and lattice temperature are provided in Fig. 5. The vertical dashed black lines indicate the initial foil boundaries. Figure 5 (a)-(c) shows the case of average lattice heating to 1.25 eV / atom (corresponding to a fluence of 3400 Jm⁻²). This is considered to be the cavitation threshold for the 300 nm simulations, as Fig. 5 (a) and (b) clearly show void formation by 130 ps. Increasing the level of heating further results in the growth of super-critical voids and subsequent ablation of the foil. Comparing the density plot to the pressure plot in Figs. 5 (a) and (b), the location of the void coincides with the peak tensile stress. The asymmetry in the heating is apparent in the space-time plot of the lattice temperature, where the irradiated side of the foil is hotter than the rear, as well as in the density profile in Fig. 5 (a) in which the rarefaction wave launched from the front surface results in a greater decrease in density relative to the rear.

Upon heating to 1.25 eV / atom the pressure value is $P=-9.43$ GPa, compared to -6.22 GPa in the 150 nm foil at the same level of heating. This is the greatest magnitude of tension of any of the 300 nm simulations, a characteristic which indicates that this level of heating corresponds to the cavitation threshold²⁵. This also coincides with the stress tensor components reaching approximately equal values. Below the cavitation threshold, at 0.95 eV / atom, the stress tensor components show slight anisotropy with σ_{xx} , σ_{yy} and σ_{zz} taking values of 6.8, 5.22 and 5.27 GPa respectively. At the cavitation threshold, the values of σ_{xx} , σ_{yy} and σ_{zz} are comparable at 9.61, 9.3 and 9.39 GPa respectively.

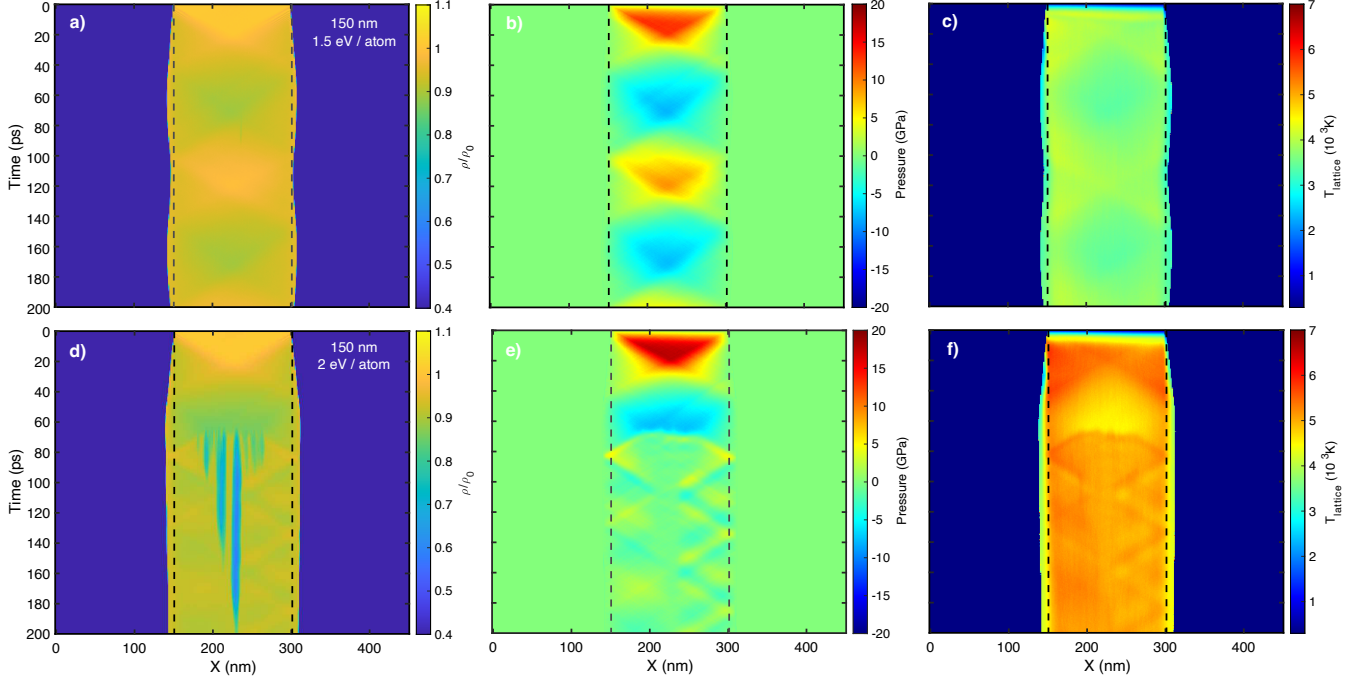


FIG. 4 Space-time plots of the density, pressure and lattice temperature for a 150 nm free-standing tantalum foil. Panels (a)-(c) correspond to average lattice heating to 1.5 eV / atom, whilst the heating is to 2 eV / atom in panels (d)-(f). The black dashed lines in each case indicate the initial foil boundaries. Panel (d) shows the formation of multiple voids within the foil as it is heated to 2 eV / atom. These voids do not reach the critical size necessary for ejection of material, and subsequently collapse, with the largest void vanishing just before the end of the simulation.

Despite the fact that a void is seeded at 130 ps, it subsequently collapses approximately 60 ps later. This behaviour is also illustrated in the phase diagram, the inset in Fig. 6. The thermodynamic trajectory in density-temperature space is plotted for different levels of heating, with the red circles corresponding to the 1.25 eV / atom case. The thermodynamic parameters are averaged over a region of four unit cells near the center of the foil. The density is calculated by summing the mass of the atoms within the slice and dividing by the sum of the per-atom volumes (the volume of the Voronoi tessellation associated with each atom). The resulting trajectory provides a macroscopic view of the system as it excludes the presence of voids. This removes the bias which could occur if the sampled region contained a large volume of voids, in which case the density of the region would be underreported (and the trajectory would penetrate deeper into the vapor dome) assuming the volume in the density calculation is that of the entire slice. The average temperature within the slice is calculated from the sum of the per-atom kinetic energies using equipartition theory. For each trajectory in Fig. 6, the color fades towards white as time progresses, with the black arrows further illustrating the passage of time. The trajectory for the 1.25 eV / atom case initially crosses the melt curve during the isochoric heating stage. It then hovers around the melt curve during the pressure release stage, moving into the vapor dome at a time of 120 ps. The reverberation waves push the trajectory back out of

the vapor dome, leaving it in a molten state just above the melt curve. This behaviour is consistent with Fig. 5 (a), where the initial void growth coincides with the atoms entering the vapor dome, and void collapse is indicated by the atoms crossing back over the vapor dome boundary and into the liquid region of the phase diagram.

It is important to note that as the molten material crosses the vapor-dome boundary it is a mixture of phases, which have different densities and temperatures. The phases are separated by analysing the Voronoi cell volumes of atoms with a density less than the value at the critical point (as reported in the Appendix). Taking the example of heating to 1.5 eV / atom, we report the gaseous phase appears at 105 ps (corresponding to the average trajectory entering the vapor-dome), with a density of 2.5 g cm^{-3} and a temperature of 4960 K. This differs from the ‘average’ properties of the mixed phase, of 14.8 g cm^{-3} and 3420 K, at the same time in the simulation. The presence of the mixed phase again highlights that destruction of the foil occurs via cavitation, as opposed to spallation which requires the sample remain in the solid phase.

As the lattice heating increases to 1.5 eV / atom (corresponding to a fluence of 3900 Jm^{-2}), the foil is again heated above the melting point. This is demonstrated in Fig. 5 (f), which shows the space-time plot of the lattice temperature. In this case it is more evident that there is a temperature gradient across the foil, with heating on the irradiated side leading to an increase in the thermal

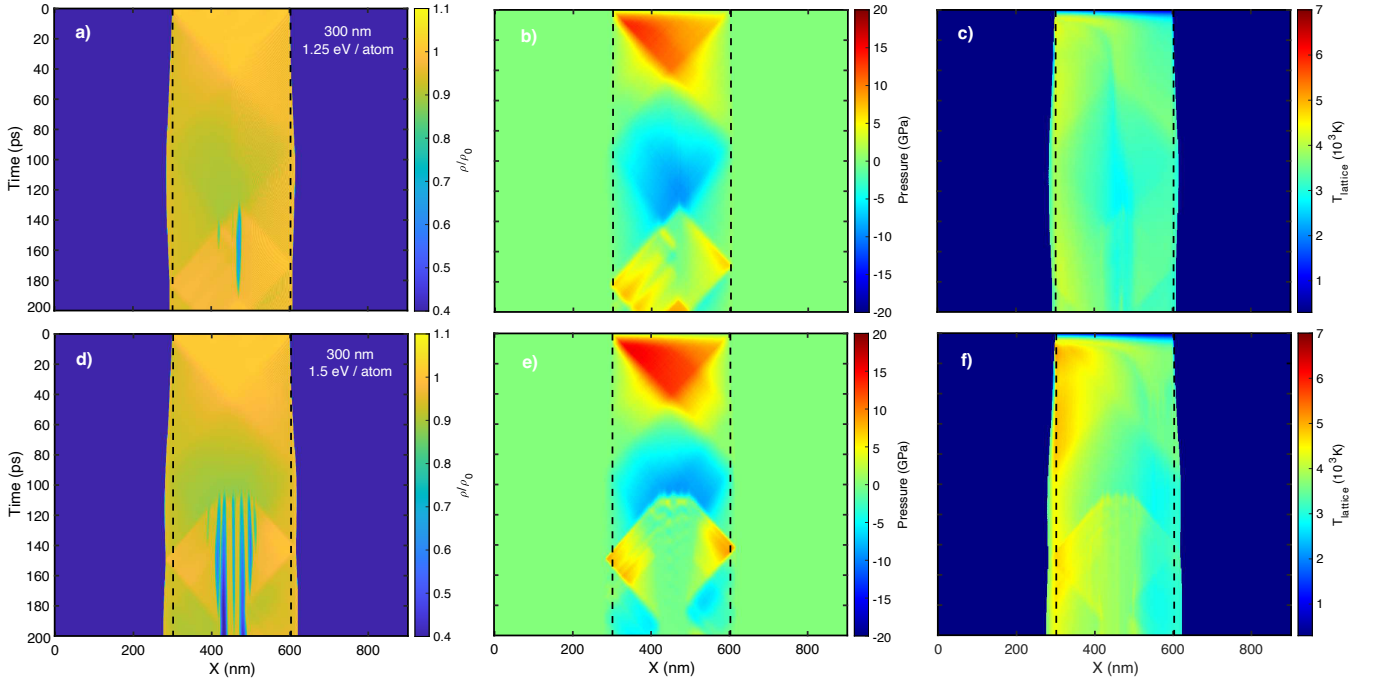


FIG. 5 Space-time plots of the density, pressure and lattice temperature for a 300 nm free-standing tantalum foil. Panels (a)-(c) correspond to average lattice heating to 1.25 eV / atom, whilst the heating is to 1.5 eV / atom in panels (d)-(f). The dashed black lines indicate the initial foil boundaries. Panel (a) demonstrates void formation and collapse upon heating of the foil to 1.25 eV / atom. As the heating increases to 1.5 eV / atom in panel (d), void nucleation occurs at an accelerated rate. Here, multiple voids reach the critical size and cause separation of material layers. Comparing panels (d) and (e) it is clear that the region of void formation coincides with the region of peak tensile stress.

pressure in this region. For this level of heating the mag-
 795 nitude of tension decreases relative to the 1.25 eV / atom
 case. The pressure reaches a value of $P = -9.33$ GPa, and
 given that the foil is molten, the components of the pres-
 800 sure tensor have similar values of σ_{xx} , σ_{yy} and σ_{zz} equal
 to 9.35, 9.38 and 9.27 GPa respectively.

Void formation occurs at later times (>100 ps) within
 the region of tension created by the intersection of the
 rarefaction waves from the front and rear surfaces. In
 contrast to the 1.25 eV / atom case, multiple voids of crit-
 805 ical size are seeded within the foil, which remain through-
 out the simulation. After void formation occurs, the pres-
 sure in the void location jumps from a negative value to
 zero. In Fig. 5 (e), it is clear that pressure reverberations
 continue within the regions of the foil which are separated
 810 by voids. Heat conduction is also stopped across these
 sections, leading to higher lattice temperature in the sec-
 tion of foil closest to the side of irradiation.

It is likely that cavitation is the ablation mechanism
 815 present in the 300 nm simulations. The phase diagram in
 the main body of Fig. 6 supports this conclusion, given
 that the phase space trajectories for the cases of heat-
 ing to 1.5 and 2 eV / atom (denoted by blue and purple
 circles, respectively) remain above the EAM melt curve
 820 at the end of the simulation. Note that for cavitation
 to occur, the trajectory must cross the melt curve, given
 that this process is described as the failure of a liquid
 under tension. Our prediction that the ablation mecha-
 nism is cavitation is then supported by the fact that for

all cases where ablation occurs, the thermodynamic tra-
 jectory does indeed cross the melt curve and enter the
 vapor dome, where voids can be seeded. In fact, for the
 cases of heating to 1.5 and 2 eV / atom, the trajectories
 remain within the vapor dome at the end of the simula-
 tion, which allows the possibility of a high rate of void
 nucleation.

In contrast to photo-thermal ablation mechanisms
 (such as vaporization), cavitation and spallation are
 driven by tensile stress. To further demonstrate that
 tensile stress plays an important role in the failure of
 the 300 nm foils, the spatial profile of the hydrodynamic
 pressure is plotted in Fig. 7. At 2 ps, following the x-
 ray pulse, the irradiated side of the foil is heated and
 consequently reaches $P \approx 5$ GPa (demonstrated by the
 blue curve). The asymmetry in the heating is evident
 by the factor of two difference in the values of hydrody-
 namic pressure at the front and rear of the foil. At 16
 ps (the orange curve) a significant region of the foil is
 still under stress confinement and continues to heat dur-
 ing the electron-lattice equilibration stage. The pressure
 has already started to decrease near the foil boundaries,
 indicating the expansion of the free-surfaces into vacuum
 and the propagation of rarefaction waves into the bulk.
 The peak tension forms at 100 ps, as indicated by the
 yellow curve, followed by void formation near the center
 of the foil. After void formation, the pressure value in
 the vicinity of the void jumps from highly negative to
 zero, as indicated in the spatial profile at 109 ps (the
 purple

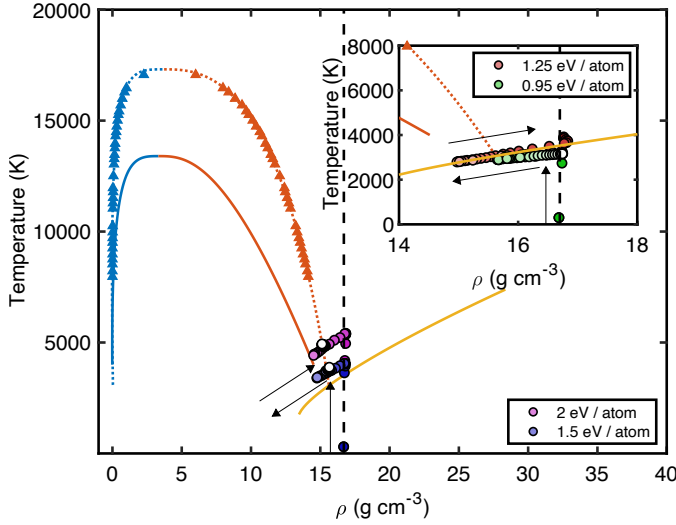


FIG. 6 Plot of the thermodynamic trajectory, averaged across a slice of length four unit cells, close to the center of the 300 nm thick foil. The density calculation excludes the presence of voids and provides a macroscopic view of the system. The phase boundaries follow the same descriptions as in Fig. 1. The purple and blue circles in the main plot are points along the thermodynamic trajectory of the central region, upon heating to 2 eV / atom and 1.5 eV / atom, respectively. The fading of the colours in the data points, and the black arrows, denote the passage of time. The inset shows the trajectory for cases where the lattice is heated to 1.25 (red circles) and 0.95 (green circles) eV / atom respectively, over a smaller region of phase space. For the lowest level of heating, the trajectory is mostly below the melt curve. This indicates cavitation is unlikely to occur given that this is defined as the failure of a liquid under tension.

curve in Fig. 7).

IV.C. 500 nm free-standing foil

As the target thickness increases to 500 nm, we again have non-uniform heating resulting in pressure and temperature gradients across the foil. This is demonstrated in the space-time plots of the pressure and lattice temperature, for the case of heating to 1.25 eV / atom (at a fluence of 5500 Jm⁻²), in Figs. 8 (b) and (c). The ablation threshold for the 500 nm simulations is, again, 1.25 eV / atom. This is confirmed in the space-time plot of the density in Fig. 8 (a) by the appearance of a critically sized void at approximately 150 ps. The late onset of void formation, relative to the thinner foil simulations, is due to the increased time it takes for the rarefaction waves from the front and rear surfaces to intersect and form tension within the foil.

Comparing the density and pressure plots in Figs. 8 (a) and (b), it is clear that the void forms in a region of tension. However, an interesting additional feature emerges, in that the region with the largest magnitude of tensile stress is spatially separated from the void formation region. The purple triangles mark the position and time at which the magnitude of tension reaches its peak. The first void appears at 150 ps, at position $X \approx 700$ nm, whereas the peak tension forms 40 ps later at a position

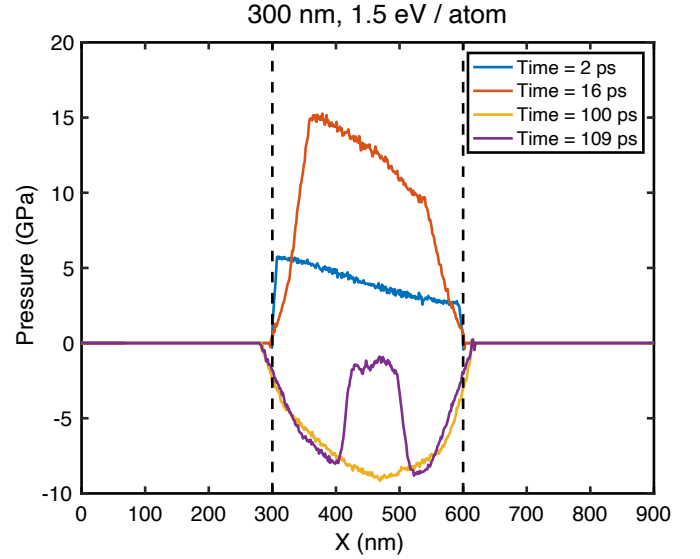


FIG. 7 Line out of hydrodynamic pressure at selected times, from a simulation with a 300 nm free-standing tantalum foil heated to 1.5 eV / atom.

of $X \approx 850$ nm. This behaviour is reported in Ref. 25 for TTM-MD simulations of ablation in nickel foils. We now confirm the same response for tantalum, via simulations of free-standing foils of greater thickness than those in Ref. 25 (although we also note that Ref. 25 reports the same behaviour in a ‘bulk’ foil, where heat conduction occurs deep into the material). The separation of the regions of peak tensile stress and void formation is the result of thermal softening. The material on the irradiated side of the foil is hotter (due to the temperature gradient) and therefore supports a weaker magnitude of tensile stress. This is relative to the rear of the foil, which is ≈ 1000 K cooler throughout the simulation, and therefore supports a larger magnitude of tension.

As the level of heating is increased to 1.5 eV / atom (corresponding to a fluence of 6650 Jm⁻²) in Fig. 8 (d)-(f), the effect of thermal softening becomes more apparent. The spatial separation between the first void and the region of peak tensile stress is increased to 219 nm, with a temporal separation of ≈ 40 ps, as indicated by the location of the purple triangle relative to the location of the first void. Whilst Fig. 8 (f) demonstrates a significant thermal gradient across the foil, another striking feature is the cooling which occurs near the center. This cooling starts around 20 ps and continues at this location until void formation begins at approximately 150 ps. We probe this region by employing additional diagnostics, including analysis of the phase space trajectory and simulated x-ray diffraction patterns. The purpose of the additional analysis is to determine the state of the material at the center of the foil, which in turn provides information on the ablation dynamics.

The positions and times captured by the additional diagnostics are marked in Fig. 9 (a) by coloured triangles. First, we look at a thermodynamic trajectory in

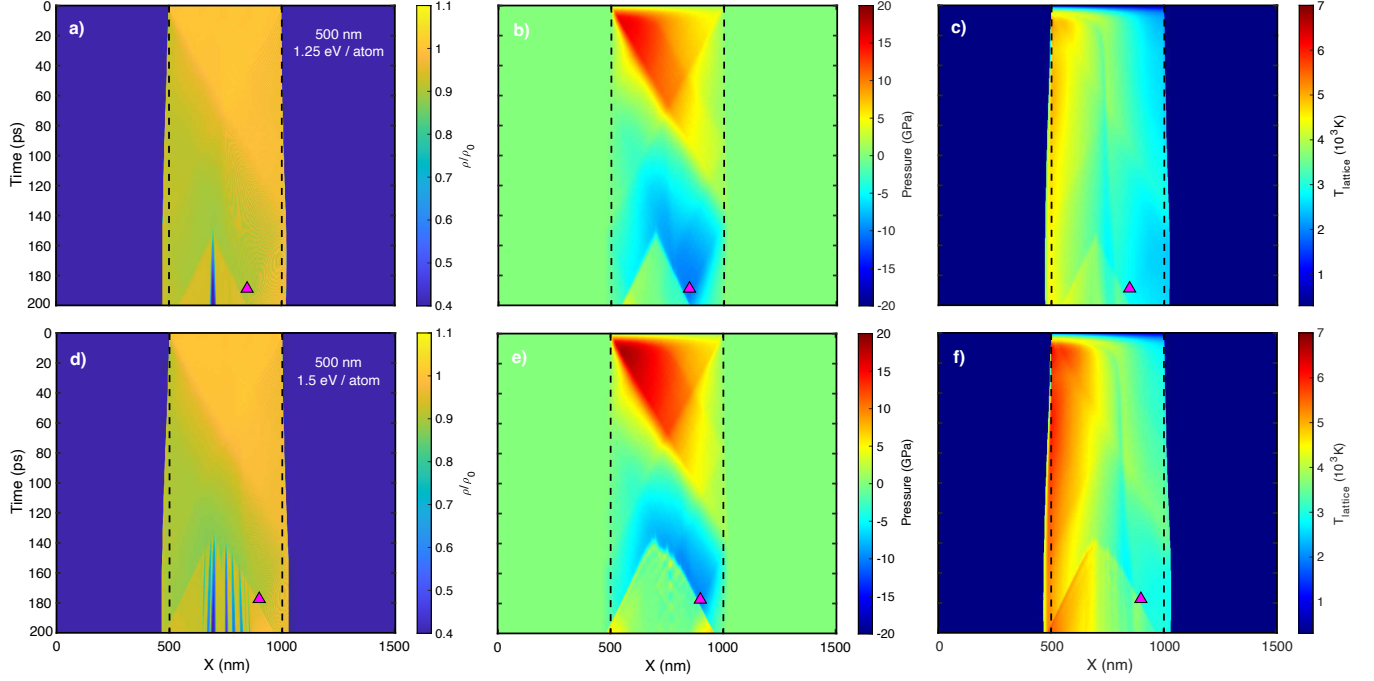


FIG. 8 Space-time plots of the density, pressure and lattice temperature for a 500 nm free-standing tantalum foil. Panels (a)-(c) correspond to average lattice heating to 1.25 eV / atom, whilst the heating is to 1.5 eV / atom in panels (d)-(f). The dashed black lines in each case indicate the initial foil boundaries, whilst the purple triangles mark the position and time at which the tensile stress reaches its maximum magnitude. Panels (a) and (d) indicate that heating to ≥ 1.25 eV / atom is sufficient to induce cavitation, with voids appearing and remaining throughout the simulations.

density-temperature space, which is representative of the macroscopic state of the system (corresponding to the average density and temperature of the mixed phase).⁹⁴⁵ This is plotted as a function of time at a constant position ($X=814.4$ nm) in Fig. 9 (b). The 500 nm thick foil is heated under the condition of stress confinement to approximately 3800 K, which is above the melt curve for the EAM potential. The peak lattice temperature occurs⁹⁵⁰ at 30 ps, when the lattice and electron subsystem reach equilibrium. The sampled volume remains stress confined at 50 ps, after which time the thermal pressure begins to release. During the subsequent cooling the trajectory remains just above the EAM potential melt curve. At 100⁹⁵⁵ ps, the rarefaction waves from the free surfaces begin to intersect, with the associated decrease in density pulling the thermodynamic trajectory into the vapor dome. At 150 ps, voids begin to form around the sampled region due to high tensile stress. This void formation causes⁹⁶⁰ release of tension and an increase in density as the trajectory propagates towards the liquid region of the phase diagram. The subsequent pressure reverberations cause the trajectory to oscillate between the liquid region of the phase diagram and the vapor dome towards the end of the simulation. We note that whilst the ‘average’ pa-⁹⁶⁵ rameters of the mixed phase are plotted in Fig. 9 (b) it is also possible to comment on the gaseous phase which appears at 150 ps. The density of this phase is 1.79 g cm^{-3} at this time, and the temperature is 7900 K.

This behaviour is also reflected in the simulated x-ray

diffraction (XRD) patterns in Fig. 9 (c). The XRD data is obtained using a custom package in LAMMPS, with further details provided in the Appendix. The XRD calculation samples a volume of $15 \times 15 \times 15$ unit cells at locations centered on the coloured triangles in Fig. 9 (a). The black diffraction pattern corresponds to ambient conditions ($t=0$ ps, with the initial lattice equilibrated to 300 K), where it is clear that there are strong peaks associated with the labelled Miller planes. The ambient peaks are still visible at 30 ps (the end of the TTM stage) and at 50 ps, although the intensity of the signal is weaker, consistent with melting of the lattice. At 100 ps and 150 ps (plotted in the inset), the only significant peak is from the (110) planes. Again, there is strong evidence of peak broadening, and the position of the peak has shifted compared to the ambient XRD pattern, suggesting the foil is under tension. Given the broad liquid background, we suggest that the cooling at the center of the foil does not result in a transition back to the solid state. The XRD patterns are in agreement with analysis of the thermodynamic trajectory, indicating the foil remains above the melt curve.

Finally, we comment on the ablation mechanism for the 500 nm foils. Fig. 8 (d) demonstrates that, upon heating to 1.5 eV / atom, there is an increased rate of void nucleation, which could be indicative of explosive boiling. However, visualization of the atomic positions reveals that the boundary between the hot material and vacuum remains sharp. In contrast, it is reported in Refs.

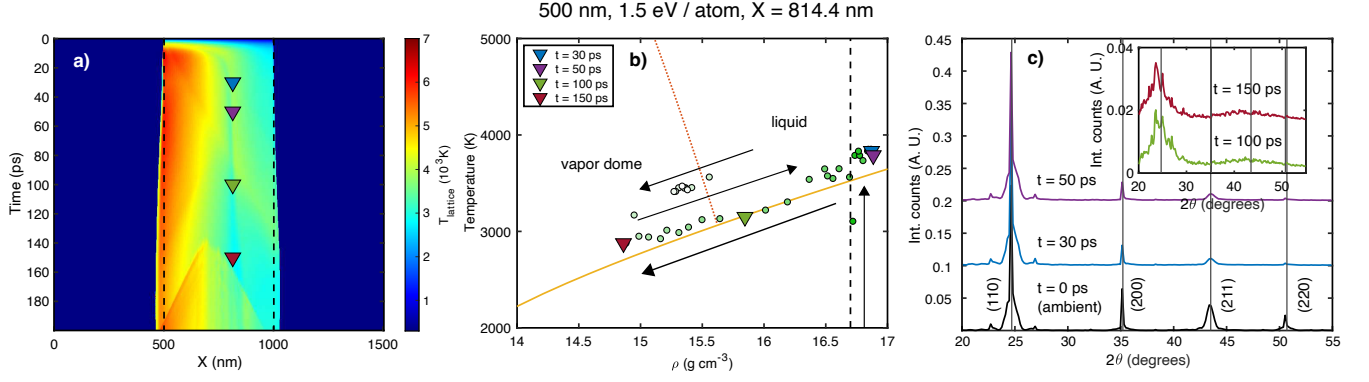


FIG. 9 (a) The space-time plot of the lattice temperature for the 500 nm-thick foil, heated to 1.5 eV / atom, is supplemented with additional diagnostic data. The triangles in panel (a) indicate the positions and times at which additional data are obtained. The thermodynamic trajectory in density-temperature space for a region of length four units cells, centered on position $X=814.4$ nm (at times corresponding to the times indicated by the coloured triangles), is plotted with green circles in panel (b). The phase boundaries are the same as those described in Fig. 1 and the black arrows indicate the passage of time. Synthetic x-ray diffraction profiles are plotted in panel (c). The black profile is the diffraction pattern at ambient conditions, whilst the colours of the additional profiles correspond to the coloured triangles in panel (a), indicating the position and time at which the data is obtained. The diffraction patterns at 100 and 150 ps are plotted in the inset with an adjusted vertical scale to emphasize the broad liquid peaks. The vertical lines in panel (c) denote the 2θ diffraction angles associated with reflection from the Bragg planes indicated by the (h,k,l) indices marked at the bottom of each line. The broad XRD profiles suggest that the crystal remains molten, despite the cooling which occurs near the center of the foil.

25 and 69 that explosive boiling is characterized by the decomposition of the material-vacuum interface into fluid₀₀₅ clusters, surrounded by gaseous atoms. This conclusion, that the ablation mechanism is cavitation for the 500 nm simulations, is supported by the preceding analysis of the thermodynamic trajectory. Even at high fluence values, the transition across the binodal line occurs well below₀₁₀ the critical point. As discussed in Section II, explosive boiling typically occurs when the material is heated close to the critical temperature.

IV.D. 1000 nm free-standing foil

The thickest foil under consideration in this study is 1000 nm, corresponding to the penetration depth for a 4.7 keV photon in ambient density tantalum. Given the increased thickness, the time required for the counter₁₀₂₀ propagating rarefaction waves from the front and rear surfaces to intersect increases, from approximately 150 ps for the 500 nm thick foil, to 250 ps in the 1000 nm thick case. The simulation run time is increased accordingly, from 200 to 400 ps.

The density, pressure and temperature profiles are plotted in Fig. 10 (a)-(c) for heating to 1.25 eV / atom (corresponding to a fluence of $11,000 \text{ Jm}^{-2}$), which is the cavitation (and ablation) threshold. It is important to highlight that this threshold is the same as for the 300₀₃₀ nm and 500 nm foils. The reason for the consistency in the behaviour is that these target thickness values exceed the distance over which the heat is carried through the target. This leads to a temperature gradient across the foil, however, the ratio of the absorption depth to the₀₃₅ target thickness is maintained at 1:1 and so the gradient is comparable in all cases. In contrast, whilst the same

1:1 ratio is present in the 150 nm foil, thermal conductivity equalizes the front and rear temperatures and there is no thermal or pressure gradient across the foil. Failure of the 1000 nm foil upon heating to 1.25 eV / atom is demonstrated in Fig. 10 (a), with a critically sized void forming at $X=1375$ nm after 225 ps. The location of the peak magnitude of the tensile stress (indicated by the purple triangle) is spatially separated from the location of the first void appearing in the foil, due to thermal softening. The pressure profile also demonstrates that stress confinement is satisfied across a larger volume of this thicker foil, with approximately no pressure release due to free-surface expansion until at least 30 ps.

Figures 10 (d)-(f) correspond to heating to 2 eV / atom, where the rate of void nucleation is seen to increase with the level of heating. This is evinced by the fact that at least eight voids appear throughout the bulk of the foil upon heating to 2 eV / atom, whereas only two voids are present at 1.25 eV / atom. This suggests thermal softening plays an important role, where the ability to support a smaller magnitude of tensile stress triggers failure multiple times within the foil. Some of these voids are seeded with insufficient energy to grow over time, whilst others merge with neighbouring voids and achieve the critical size.

In Fig. 10 (f), we see that the lattice temperature in the void formation region exceeds the boiling point predicted for the EAM potential. Closer inspection of the voids shows that they contain very few gaseous atoms and there is still a clear boundary between regions which are separated by the formation of voids. This suggests that ablation is driven by mechanical means up to at least the boiling point, and that the effects of thermal processes such as explosive boiling are negligible even

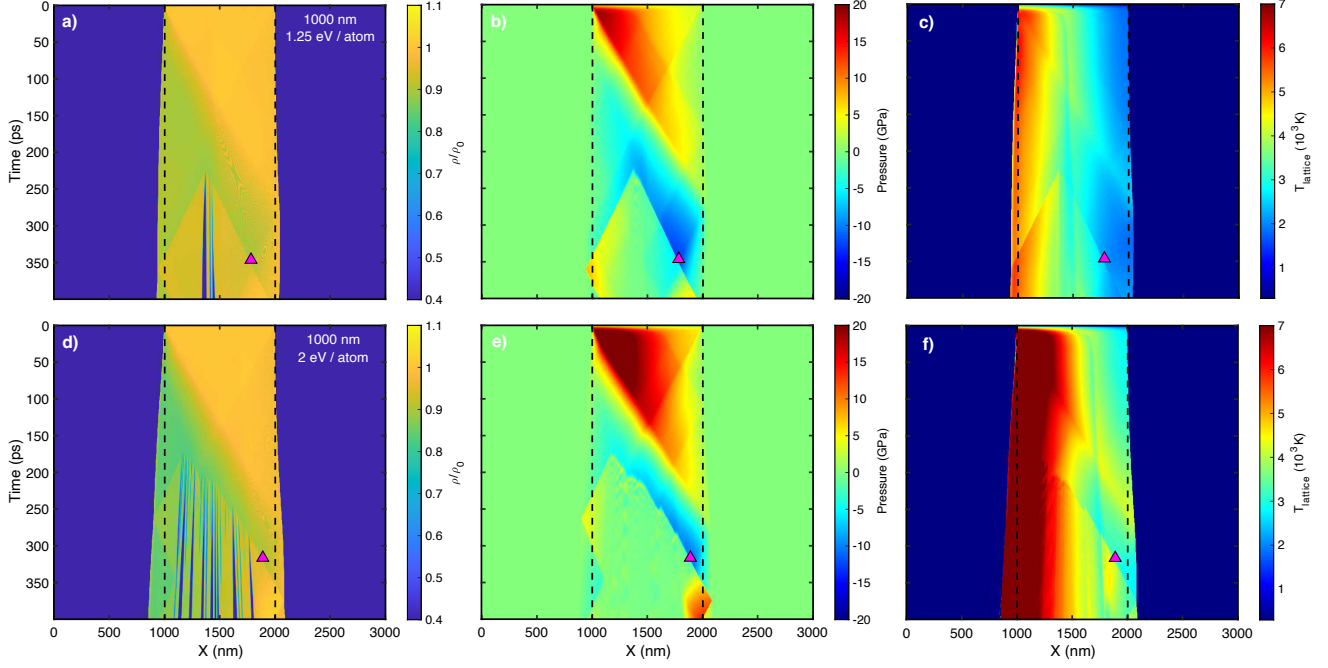


FIG. 10 Space-time plots of the density, pressure and lattice temperature for a 1000 nm thick free-standing tantalum foil. Panels (a)-(c) correspond to average lattice heating to 1.25 eV / atom, whilst the heating is to 2 eV / atom in panels (d)-(f). In panel (d) the increased rate of void nucleation is evident. The dashed black lines in each panel indicate the initial foil boundaries and the purple triangles denote the position and time at which the tensile stress reaches its maximum magnitude. Comparing panels (a) and (b), it is clear that the void formation region is separate from the region of peak tensile stress, as a result of thermal softening.

at this high level of heating (to 2 eV / atom). This is also consistent with the thermodynamic trajectories in density-temperature space, which show ablation occurs when the trajectory enters the vapor dome. As noted in Section II, it is generally understood that phase explosion requires heating to approximately 90% of the critical temperature. Analysis of the phase space trajectory reveals that even for the front surface of the foil, where the temperature is highest, the trajectory enters the vapor dome at a temperature of 10^4 K, well below the critical point.

Finally, the spatial profile of the hydrodynamic pressure is plotted in Fig. 11, for selected times within the simulation. This plot highlights the presence of the extreme thermal and stress gradients across the 1000 nm thick foil. Between the times of 2 ps and 30 ps, the foil continues to heat due to the slow transfer of energy from the electrons to the lattice. It is also clear that stress confinement occurs over a large volume of the foil, with only a small degree of pressure release from the irradiated surface at 30 ps. The tension on the irradiated side of the foil exceeds the tensile strength of the material around 170 ps, subsequently followed by void formation as indicated by the black arrow. This profile is demonstrably different to that of the 300 nm foil, presented in Fig. 7. For the 1000 nm foil, we see void formation begins close to the irradiated surface and propagates into the bulk, where the magnitude of tensile stress is greater. At 220 ps, we can see the tension is still increasing in magnitude towards the rear of the foil. This demonstrates the

important role thermal softening plays for the thick foil, whereas for the 300 nm foil in Fig. 7 the magnitude of the peak tensile stress coincides with the void formation region.

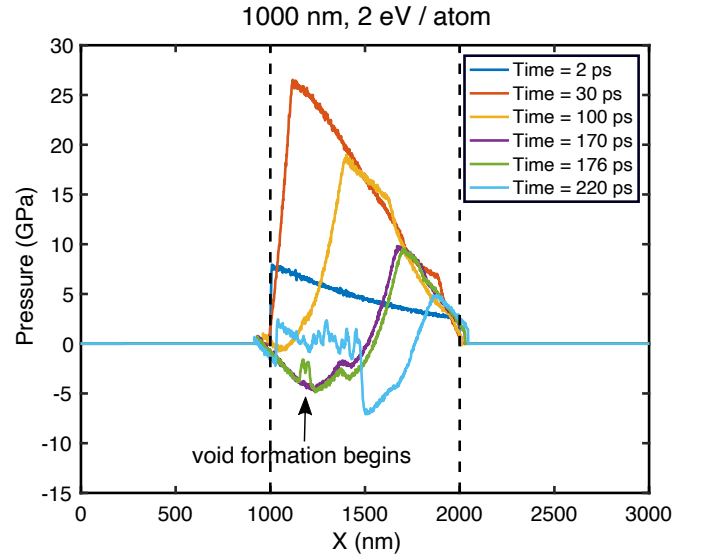


FIG. 11 Line out of the hydrodynamic pressure at selected times, from a simulation with a 1000 nm thick free-standing tantalum foil heated to 2 eV / atom. The black arrow indicates the position of the first void, which appears in the simulation at 180 ps.

V. DISCUSSION

The aim of this investigation is to determine the cavitation threshold and subsequent ablation dynamics for tantalum foils over a range of thickness values. We report that the cavitation threshold for the 150 nm foil is 2 eV / atom, which is higher than that of the thicker foils. As the foil thickness increases to ≥ 300 nm, the behaviour becomes consistent and we observe the cavitation threshold at 1.25 eV / atom, independently of the foil thickness. In all of the simulations, we confirm that destruction of the foil results from cavitation and subsequent ablation, indicating that damage is mechanical in nature over the range of incident x-ray energies investigated. Evidence of cavitation is presented in the form of space-time plots, which show the formation and growth of voids coincident with regions where the magnitude of tensile stress is large. Additional analysis of the trajectory the foils follow through (density-temperature) phase space shows the subsequent stages of isochoric heating across the melt curve, followed by a transition across the coexistence line into the vapor dome where cavitation is increasingly likely.

To further rule out the presence of explosive boiling in the simulations, the upstream free surfaces of the foils were analyzed, which are subjected to the highest temperatures in the sample. Despite the extreme temperatures (the free surface reaches 10,300 K for the 1000 nm foil heated to 2 eV / atom) we observed no signatures of vaporization. We report no surface removal of atoms or void formation within the first 100 nm of the foil, for any conditions studied, up to several hundred ps after irradiation. The free surfaces remain well defined and show free surface velocities (of the order several hundred ms^{-1}) consistent with those expected on the basis of hydrodynamic release of fully dense Ta⁷⁰. The average thermodynamic trajectory of the material located within four units cells of the upstream free surface remains close to the liquid-vapor dome binodal, due to the absence of tensile stress in this region. This is consistent with a lack of significant vaporization, indicating primarily mechanical ablation for the simulation parameters chosen in this study.

We now provide an overview of the simulation results, in terms of the maximum and minimum values of the hydrostatic pressure (P), and compare against existing studies. Figure 12 (a) shows the peak pressure for each foil (denoted with a different colour) as a function of the deposited energy, measured in eV / atom. It is important to note that, here, the position of the peak pressure varies with the foil thickness and the level of heating. For each thickness under consideration, the peak pressure increases linearly with the degree of heating. This behaviour is expected, given that this reflects the increasing thermal pressure due to stress confinement over a larger region of the foil. As described in Ref. 25, the condition for stress confinement is that the duration of the radiation pulse and the electron-lattice equilibration

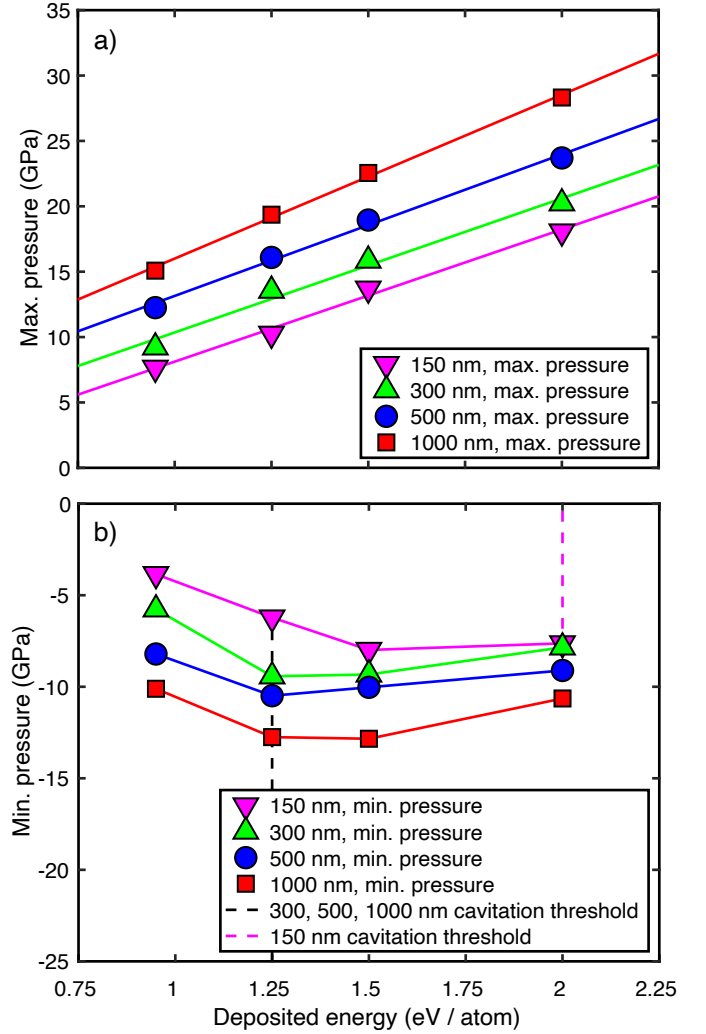


FIG. 12 Plots of the (a) maximum hydrostatic pressure and (b) the hydrodynamic pressure at the time of maximum tension (i.e. peak σ_{xx}) for different levels of lattice heating. Each colour corresponds to a different foil thickness, with purple, green, blue and red indicating foils of 150, 300, 500 and 1000 nm thickness, respectively. The dashed purple line in (b) indicates the cavitation threshold for the 150 nm foil, whilst the dashed black line is the ablation threshold for the thicker foils.

time must be less than the timescale associated with thermal expansion (related to the speed of sound at ambient density). For the 150 nm foil, at the ablation threshold, the time scale associated with thermal expansion is approximately 40 ps. This is comparable to the electron-lattice equilibration time of 30 ps. As the foil thickness increases to 1000 nm, the thermal expansion time scale increases to approximately 227 ps, which is much greater than the equilibration time (which is still 30 ps). This means a larger volume of the foil is heated under the condition of stress confinement, resulting in higher thermal pressure.

In Fig. 12 (b) the hydrostatic pressure at the time of maximum tension, which we refer to as the minimum pressure, is plotted for each foil thickness as a function

of deposited energy. Again, the position of the measurement varies with the target thickness and level of heating. As the thickness increases, the peak tension shifts towards the rear of the foil due to thermal softening. As discussed, the magnitude of the minimum pressure is lower for the 150 nm foil (indicated by the purple curve) due to poor stress confinement. Although thermal softening is present for all of the simulations, the pressure release in the 150 nm foil is insufficient to exceed the tensile strength of the material until the heating reaches 2 eV / atom. The magnitude of the minimum pressure reached in the 150 nm simulations therefore increases with deposited energy, indicating the release of a larger thermal pressure at the end of the isochoric heating stage. For the thicker foils (≥ 300 nm), the importance of thermal softening can be inferred from the shape of the curves in Fig. 12 (b). At the lowest level of heating, to 0.95 eV / atom, thermal softening is insufficient to lead to failure of the foil. Since the tensile strength has not been exceeded, marginally increased magnitudes of minimum pressure can be withstood upon further heating. The cavitation threshold for each foil thickness is then located at the minimum of the corresponding curve in Fig. 12 (b).

As the degree of heating increases, so does the magnitude of thermal softening, reducing the maximum tensile stress which can be supported within the foil. Note that the curve corresponding to the 150 nm foil follows the same trend as the thicker foils, given that the cavitation threshold is located at 2 eV / atom. It is expected that for further heating the magnitude of the minimum pressure would decrease, as it does for the thicker foils. It should also be noted that this behaviour, in which the magnitude of the peak tensile stress increases with fluence up to the cavitation threshold before either saturating or decreasing, is in excellent agreement with the simulation results presented in Ref. 25 for nickel foils.

The hydrodynamic pressure values in Fig. 12 (b) are measured at the location and time where the magnitude of tension reaches its maximum, which varies with foil thickness and level of heating. To obtain an estimate of the tensile strength of tantalum under extreme heating conditions, we must also determine the magnitude of tension at the point of failure. Such analysis also enables comparison against recent simulation²⁷ and experimental studies^{39,71} investigating the tensile strength of warm dense tantalum, which will be detailed later. In Fig. 13 (a) the minimum pressure value at the point of failure is plotted as a function of the level of heating, for different foil thickness values (denoted by different colored markers). The point of failure in each simulation is the location of the void closest to the irradiated side of the foil. The simulations with a higher degree of heating exhibit multiple voids which could be used for the measurement but for consistency, and to ensure we are reporting the maximum spatial separation of the void formation region from the region of peak tensile stress, we choose the void closest to the irradiated side. The measurement at the point of failure is made by analyzing the spatial pressure

profile (such as those demonstrated in Figs. 7 and 11), taking the value of pressure at the moment it begins to ‘jump’ back towards zero, following void formation. Note that the data set is reduced compared to Fig. 12 as we only include simulations where the cavitation threshold has been exceeded. As the level of heating increases, the magnitude of the minimum pressure at the point of failure decreases. The effect becomes more pronounced as the foil thickness increases, due to the increasing thermal gradient, with failure occurring in the hot material on the side of irradiation. For example, for the 1000 nm foil, the minimum pressure at the point of failure decreases in magnitude from -7.82 GPa to -3.97 GPa, as the level of heating increases from 1.25 to 2 eV / atom.

The value of the pressure minimum is plotted as a function of the normalized position within the foil where each measurement is taken in Fig. 13 (b). This normalized distance is defined as $X^* = (x_m - \ell) / \ell$, where x_m is the location of the measurement, and ℓ is the target thickness. The measurements taken at the location where the tension reaches its maximum magnitude (i.e. the same data points which appear in Fig. 12 (b)) are denoted with colored circles, with different colors representing different target thickness values. In contrast, the colored triangles indicate measurements at the point of failure.

The stars correspond to the pressure (at the time of maximum tension) from simulations where no cavitation or ablation occurs, which act as a reference for each value of target thickness. For the 150 nm foil, the uniform heating means the rarefaction waves from the free surfaces overlap at the center of the foil. As expected, the peak tension occurs at a value $X^* = 0.5$, indicated by the purple star. As the thickness increases, the asymmetry in the heating means the peak tension occurs deeper within the foil, at values of X^* tending towards unity.

The key point of Fig. 13 (b) is that, as the target thickness increases, the separation of the void formation region and the region of peak tensile stress also increases. This is evinced by the greater distance between the clusters of triangular and circular markers, for each target thickness. Furthermore, Fig. 13 (b) acts as a means of translating between Fig. 12 (b), which shows the minimum pressure as a function of the level of heating, regardless of the location within the foil, and Fig. 13 (a), where the minimum pressure is plotted as a function of heating, as measured at the point of failure.

The thermal softening effect is investigated in more detail in Fig. 14. The colored markers denote data points from this investigation, with each color corresponding to a different target thickness. These data points are compared to the results of MD simulations investigating quasi-isentropic (QI) compression and tension of tantalum in Hahn *et al.*²⁷, denoted with a dashed orange curve. The QI method is employed to replicate the conditions of shock compression and release experiments, with the advantage that such simulations can provide homogeneous compression and stretching across the sample at a fixed strain rate. The simulations in Ref. 27 are found to be

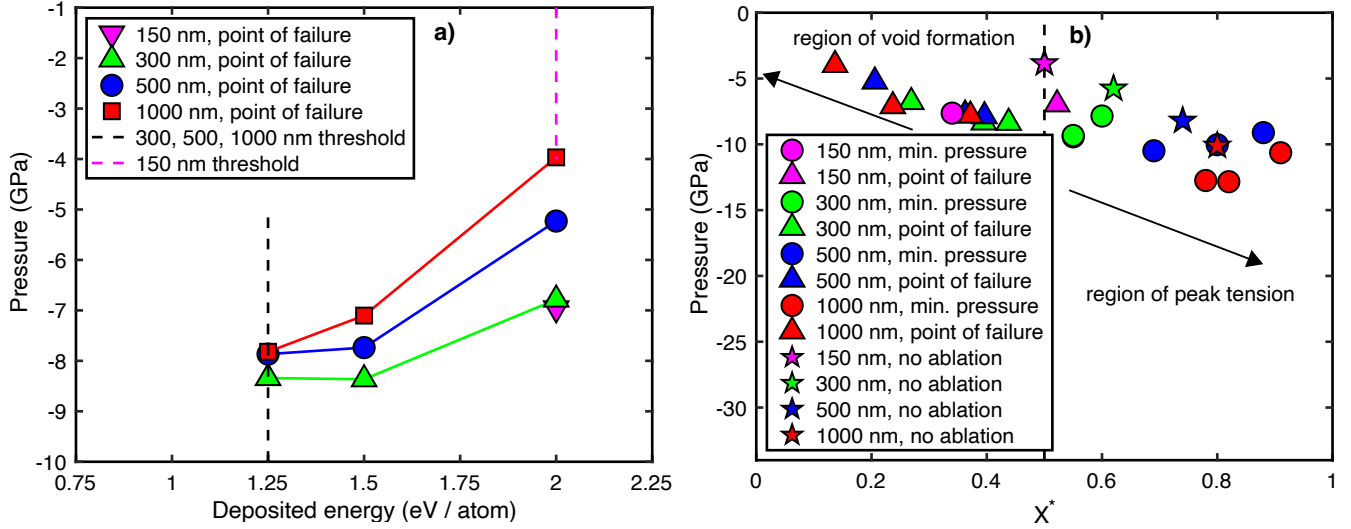


FIG. 13 (a) Pressure at the time of peak tension plotted as a function of the level of heating, for different values of foil thickness. In contrast to Fig. 12, measurements are taken at the point of failure, i.e. the location of void formation closest to the irradiated side. As the level of heating increases, the magnitude of tension within the foil decreases. (b) Pressure at the time of maximum tension, plotted as a function of the normalized position within the foil. The coloured triangles denote measurements at the point of failure, and the coloured circles indicate measurements taken at the location where the tension reaches its maximum magnitude. The coloured stars indicate cases where no cavitation or ablation occurs, serving as references, whilst the vertical dashed line marks the center of the foil. Panel (b) shows that, as the thickness increases, void formation occurs closer to the irradiated surface whilst the peak tension shifts towards the foil rear surface.

in excellent agreement with the results of the laser ablation experiment (using an optical laser pulse) in Ashitkov *et al.*⁷¹, as indicated by the orange diamond. We further relate our results to the experimental and ab-initio MD (AIMD) data points presented in Katagiri *et al.*³⁹, denoted by black circles with corresponding error bars. Whilst Ref. 39 is not strictly a laser ablation experiment, it does report on the formation of tension within liquid tantalum during a hydrodynamic compression and release experiment, and predicts the scaling of the magnitude of tension as a function of temperature, which is also an aim of this investigation. The grey curve in Fig. 14 is the EAM potential melt curve, extrapolated to negative pressures. All of our data points sit above the melt curve, again indicating that the foil must first melt before failure can occur via cavitation. Finally, we include the predictions of classical nucleation theory. This is plotted with a solid blue curve in Fig. 14 using data from Ref. 39.

Figure 14 demonstrates that the trend in our data is in agreement with Ref. 27. The QI compression and tension in those simulations is performed at a constant strain rate of $\dot{\epsilon} = 10^9 \text{ s}^{-1}$. Estimation of the peak strain rate from our simulations predicts values in the range $\dot{\epsilon} \approx 1\text{--}3 \times 10^9 \text{ s}^{-1}$, depending on the thickness and level of heating, hence we expect the ablation dynamics to be similar given we are probing a similar region in strain-rate space. We note that the magnitude of tension is somewhat higher in this study compared to the data in Ref. 27. This could be due to the choice of potential, as Ref. 27 uses the Ravelo Ta1 potential³¹ which has slightly

different thermal properties (such as underestimating the thermal expansion coefficient to a greater degree than the Ta2 potential). Another consideration is that the QI compression and tension in Ref. 27 leads to a high density of dislocations, which is predicted to reduce the tensile strength of the material, which may again explain the higher magnitudes of tension in our investigation.

We also find agreement with Ref. 39, whose data points also sit somewhere between the simulations of Ref. 27 and the predictions of classical nucleation theory. Similarly to Ref. 39, our data points sit above the curve predicted by classical nucleation theory. This discrepancy could be caused by anisotropy during the pressure release, as highlighted in Ref. 39. The agreement between the data sets in Fig. 14 indicates that our simulations are probing a similar region of P-T space as the aforementioned experiment. It is reasonable then that TTM-MD simulations may be used as a means of interpreting the results of hydrodynamic compression and release experiments, and understanding the underlying physics on time scales which the experiment cannot probe (i.e. fs resolution of the temperature evolution and ps resolution of atomic trajectories).

It is important to state that we are reporting agreement between the results of TTM-MD simulations (this study), MD simulations of quasi-isentropic compression and tension (Ref. 27), and experimental and AIMD data points from a hydrodynamic experiment (Ref. 39). This agreement suggests an equivalence between hydrodynamic compression and release experiments and isochoric heating experiments which, to our knowledge, has

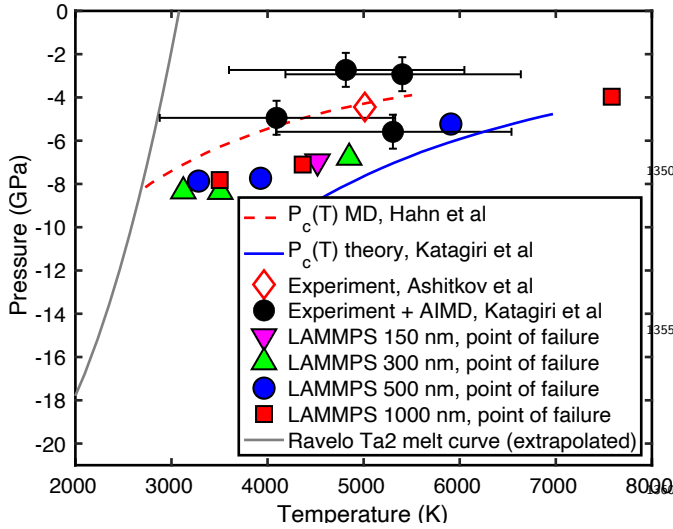


FIG. 14 Demonstration of the thermal softening effect by which the magnitude of the tensile stress within the foil is reduced upon heating. Each measurement is taken at the point of failure, i.e. at the location of void formation closest to the side of irradiation. The purple, green, blue and red markers correspond to data points in this investigation, with each colour denoting a different target thickness. We compare against the results of quasi-isentropic compression and tension simulations in Hahn *et al.*²⁷, indicated by the dashed orange line. We also compare the data points to the experimental and AIMD data presented in Katagiri *et al.*³⁹, represented by the black circles with associated error bars. Following Ref. 39, we also include a data point from the laser ablation experiment of Ashitkov *et al.*⁷¹, and the prediction of nucleation theory, denoted by the solid blue curve, plotted using data from Ref. 39. The grey curve is the melt curve for the Ravelo Ta2 potential³¹, extrapolated to negative pressures using the fitting equation presented in Ref. 50 and converted to density-temperature space using the EoS spreadsheet in Ref. 51

not yet been discussed in the literature.

This result has implications for the interpretation of existing experiments and in guiding future studies. The relevance to hydrodynamic experiments probing liquid tantalum states has already been discussed, but the simulations can also improve understanding of investigations into the tensile strength of solid tantalum. For example, the spall strength of tantalum is investigated experimentally in Ref. 72, and supported by large-scale MD simulations. In those simulations, a piston is launched into the sample to recreate conditions of hydrodynamic compression and release. The temperatures are likely much lower in Ref. 72 compared to our investigation, given that failure occurs in the solid state. However, the reported spall strength of ≈ 15 GPa from a 3950 nm thick sample in Ref. 72 is comparable to the values of tension in our study, for thick foils heated to below the threshold for cavitation (for example, see the values of the minimum pressure for the 1000 nm foil in Fig. 12 (b)). Although there appears to be agreement between the experiment and simulations at low temperatures, the TTM-MD simulations provide a means of probing the tensile strength of materials at extreme temperatures, perhaps even outside the P-T regime currently accessible at XFELs.

VI. CONCLUSIONS

Much prior research into the behaviour of tantalum at extreme conditions has focused on computational investigations of its tensile properties^{27,31,73}, such as the spall strength and stress induced phase transitions³⁰. Given that facilities are now capable of delivering exceptionally bright pulses of high energy x-rays, there is a desire to investigate the ablation processes which underpin the destruction of (micron) thick tantalum foils under high energy x-ray irradiation. Whilst these mechanisms remain poorly understood within the high energy x-ray regime, progress in understanding ablation under optical and soft x-ray pulses has historically been facilitated by computer simulations. Specifically, these simulations employ hybrid two-temperature MD codes, which are capable of describing the ablation process on the atomic scale and on ps time frames.

In this study, the cavitation threshold and ablation dynamics are investigated for a range of tantalum foil thicknesses and under different levels of heating. The x-ray energies vary in the range 1-5 keV, which is at the lower end of the operating energy range of current XFEL facilities. We reiterate that stress confinement occurs over a small volume for the thin (150 nm thick) foil, resulting in a lower thermal pressure, and a smaller magnitude of tension upon release of this stress confined state. The cavitation threshold for this case is 2 eV / atom, which is greater than that of the thicker foils. As the thickness increases to ≥ 300 nm, we report a cavitation threshold of 1.25 eV / atom, which is independent of the foil thickness. These results are supported by the experimental findings in McHardy *et al.*⁷⁴, in which tantalum foils irradiated at the EuXFEL (at an energy of 17.8 keV) exhibit damage patterns consistent with the ejection of liquid material from the laser spot, and clear shearing of the foils suggests tensile stress plays a role in the ablation process.

This current investigation aims to elucidate the time scale over which x-ray energy is coupled into the electrons and the lattice immediately following irradiation, and to determine the damage mechanism. In this respect, we find that electron-lattice equilibration occurs over a time scale of approximately 30 ps, and damage occurs due to cavitation and subsequent ablation. The results of this study are timely in that they extend TTM-MD simulations into the regime of high energy x-rays, assisting in the interpretation of experimental results. Improved understanding of the underlying mechanisms of energy transfer and ablation during high energy x-ray irradiation may lead to applications including micro-structuring of material surfaces and development of beam-line optics for free-electron lasers.

Finally, we note some limitations of the current investigation. As previously stated, the upper limit of the incident x-ray energy is restricted to 5 keV. This is within the operating range of XFEL facilities, and can still be used to describe the behaviour observed in in XFEL experi-

ments. However, the delivered x-ray energy is lower than the 17.8 keV typically delivered by the EuXFEL. The restriction arises because, as the x-ray energy increases, ballistic electrons are expected to play a more important role in the interaction. More precise modelling of these ballistic electrons, in terms of their energy distribution and mean free path, is needed before these can be implemented in the TTM-MD simulations. It is also worth noting that the simulations do not consider ionization processes, or account for plasma behaviour, which are important at higher fluences. Follow-up work should address these limitations, possibly by using Monte-Carlo simulations to predict the ballistic electron range. We also note that the electron thermal conductivity chosen in this study is likely an underestimate of the true value, however progress in this respect will involve turning to DFT calculations to predict the functional form of the electron thermal conductivity with temperature, and modifying the simulation source code to incorporate this. We also note that we have used idealized tantalum crystals in this investigation, constructed from a regular bcc lattice. Under experimental conditions, it is often necessary to consider the role of defects in the crystal lattice, which may influence its mechanical properties. Future work may address the role of defects on the determination of the ablation threshold in more detail, however we direct the interested reader to Refs. 27, 50, 75, and 76, where the spall strength of tantalum has already been investigated as a function of grain size and strain rate.

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APPENDIX

Mixed phase simulations

Mixed phase simulations were performed to determine the vapor dome boundaries for the Ravelo Ta2 potential³¹ used in this study. The simulation scheme involves first heating a tantalum lattice to a target temperature, in the range 8×10^3 - 17×10^3 K. The system is given time to

evolve under a NPT barostat with a target pressure of zero bar, over a typical time scale of 50 ps. This generates a stable sample of molten tantalum. The simulation box is then resized to accommodate regions of vacuum on either side of the (molten) liquid sample, into which atoms of the gaseous phase can evaporate. The system is given a further 50 ps for the vapor to saturate, then the liquid and vapor phases evolve according to a NVT thermostat until a stable state is reached. This stable state is defined as one in which the temperatures in the liquid and vapor phases differ by no more than 50 K, and the vapor density does not vary by more than 1% in a 25 ps period. For the system to properly thermalize, we impose the condition that the size of the vacuum region (and therefore of the vapor phase) must be greater than the mean-free-path of an atom in the vapor phase. Note that the requirement that the vapor phase reaches a stable state restrains the range of temperatures over which the mixed phase simulations can be employed. Below a temperature of 8×10^3 K, the typical collision time between atoms exceeds 500 ps, making it unlikely that the liquid and vapor will reach thermodynamic equilibrium over the time scales accessible in molecular dynamics simulations. We also note that in the case where the saturated vapor phase contains fewer than 1000 atoms, the dimensions of the simulation box are expanded to accommodate more atoms of this phase, and the system is given time to relax to a stable state under the NVT thermostat, as described above. These steps (evaporation and relaxation) are iterated until a sufficient number of atoms exist in the vapor phase.

After reaching equilibrium, the average density profile is constructed by sampling the system at regular intervals over a time scale of 250 ps. Given a sufficient number of atoms in the initial tantalum lattice, the liquid region remains stable and a clear distinction between the liquid and vapor regions is observed (albeit separated by a thin transition layer). The density spatial profile is then fitted with a hyperbolic tangent function, giving the density of the liquid and vapor phases at the target temperature. Repeating this simulation scheme over a range of target temperatures enables the boundary of the vapor dome to be mapped out. The phase boundaries obtained from the mixed phase simulations are plotted in Fig. 1, where the dotted blue and orange curves are obtained by fitting the simulation data points to the following equations (adapted from Ref. 48):

$$\rho_{\text{vapor}} = \rho_{\text{diam}} - \rho_{\text{diff}} \quad (6)$$

$$\rho_{\text{liquid}} = \rho_{\text{diam}} + \rho_{\text{diff}} \quad (7)$$

where

$$\rho_{\text{diam}} = \rho_c \left(1 + a(T_c - T) + c(T_c - T)^{\frac{2}{3}} \right) \quad (8)$$

$$\rho_{\text{diff}} = b(T_c - T)^{\frac{1}{3}} \left(1 + b_2(T_c - T)^{\frac{2}{3}} \right) \quad (9)$$

with $T_c = 17.3 \times 10^3$ K and $\rho_c = 3.64$ g cm⁻³ denoting the critical temperature and density, respectively, and

the remaining fitting parameters detailed below.

$$a = -5.12 \times 10^{-5} \text{ K}^{-1} \quad (10)$$

$$b = 0.384 \text{ g cm}^{-3} \text{ K}^{-1/3} \quad (11)$$

$$b_2 = -2.77 \times 10^{-4} \text{ K}^{-2/3} \quad (12)^{565}$$

$$c = 0.00320 \text{ K}^{-2/3} \quad (13)$$

Comparing the mixed phase simulation results to the phase boundaries reported by Leitner *et al.* (denoted by the solid blue and orange curves in Fig. 1), it is clear that the mixed phase data points sit at higher temperatures, and predict a higher critical temperature. This discrepancy could be explained by the fact that the Ravelo Ta2 potential underestimates the Grüneisen parameter at ambient conditions by approximately 40% (as reported in Ref. 33), which subsequently results in an underestimate of the thermal expansion coefficient of tantalum. For a given temperature, we would then expect the simulations to predict a higher density than the experimentally measured value, which is indeed the case. A potential consequence of this higher critical temperature compared to the experimental measurement is that the simulations will overestimate the threshold for high fluence ablation mechanisms. Whilst this is, again, a limitation imposed on the study by the choice of potential, we reiterate that it is extremely difficult to find a potential which faithfully encapsulates all of the physical processes leading to ablation. Given the demonstrable ability of the Ravelo Ta2 potential to recreate the melt curve of tantalum, and its robustness in the high pressure regime, we believe this is a suitable choice.

Estimating the critical void radius

The critical void size is estimated using a configuration similar to that of the mixed-phase simulations. The steps are as follows:

- A tantalum slab is heated to $12 \times 10^3 \text{ K}$ under a NPT barostat with a target pressure of zero bar for 60 ps.
- The liquid slab is subjected to isotropic tension at a strain rate of $6 \times 10^{-3} \text{ s}^{-1}$ for 250 ps.
- The dimensions of the simulation box are increased to create vacuum regions into which the vapor phase can evaporate over a period of 50 ps.
- The system is given a further 800 ps for the vapor phase to stabilize (in terms of number density).
- Averages of the density spatial profile are obtained at regular intervals over a time scale of 25 ps.

Following the approach described in Ref. 43 using classical nucleation theory, the parameters which are required to estimate the critical void size are the width of the transition region between the liquid and vapor phases (denoted δ) and the value of surface tension across a planar interface of liquid and vapor (denoted γ_0). Fitting a

hyperbolic tangent function to the average density profile yields a value of 2.07 \AA for the width of the liquid-vapor interface. As described in Ref. 43, this width can be used to approximate the characteristic length in the so-called Tolman equation, relating the size of the voids to the values of tension across the interface. The above estimate of the Tolman length is in excellent agreement with the prediction of the liquid drop model in Ref. 77.

The surface tension is evaluated using the method described in Ref. 78, where the spatial profile of the differential stress (the difference between the normal and tangential components of the stress tensor) is integrated across the interface. This yields $\gamma_0 = 0.4479 \text{ Jm}^{-2}$. This is greater than the value obtained via scaling of the surface tension with temperature reported in Ref. 79, which predicts a value of 0.3 Jm^{-2} when extrapolated to a temperature of $12 \times 10^3 \text{ K}$. It is also higher than the value of 0.22 Jm^{-2} predicted by DFT in Ref. 80. The discrepancy may be related to the fact the potential underestimates the thermal expansion coefficient, resulting in higher density (and surface tension) at a given temperature compared to experimental measurements.

The critical void radius is then calculated from Eqn. (14), below⁴³.

$$\frac{1 + \frac{3\delta}{r_c} \frac{r_0^*}{r_c}}{(1 + \frac{2\delta}{r_c})^2 \frac{r_0^*}{r_c}} = 1 \quad (14)$$

The quantity $r_0^* = 2\gamma_0/|P|$ is estimated using the maximum magnitude of tension in the simulation, which is 1.33 GPa. Substituting δ and γ_0 into Eqn. (14) yields a critical void radius of $r_c = 4.03 \text{ \AA}$. This value is much smaller than the transverse dimensions of the simulation box, giving confidence that the growth of critically sized voids is not restrained by the simulation dimensions and therefore that the cavitation threshold is correctly reported.

Synthetic x-ray diffraction data

In this article we demonstrate that under hard x-ray irradiation, ablation occurs in free-standing tantalum foils upon heating to 1.25 eV / atom . The ablation mechanism, over the range of fluence investigated, is cavitation, where the tension within the molten foil exceeds the material tensile strength. The primary diagnostic for determining that the metal is molten is by identifying regions within the foil where the lattice temperature exceeds the melting point of the EAM potential used in this study. It is possible to corroborate this result using synthetic x-ray diffraction from the TTM-MD simulations, as presented in this section.

Synthetic x-ray diffraction (XRD) patterns are obtained from the simulations using the custom USER-DIFFRACTION package in LAMMPS, described in detail in Refs. 81 and 82. The XRD calculation can be performed in post-processing by loading in atomic positions from a standard dump file. This allows flexibility for the user to vary the wavelength of the incident x-ray

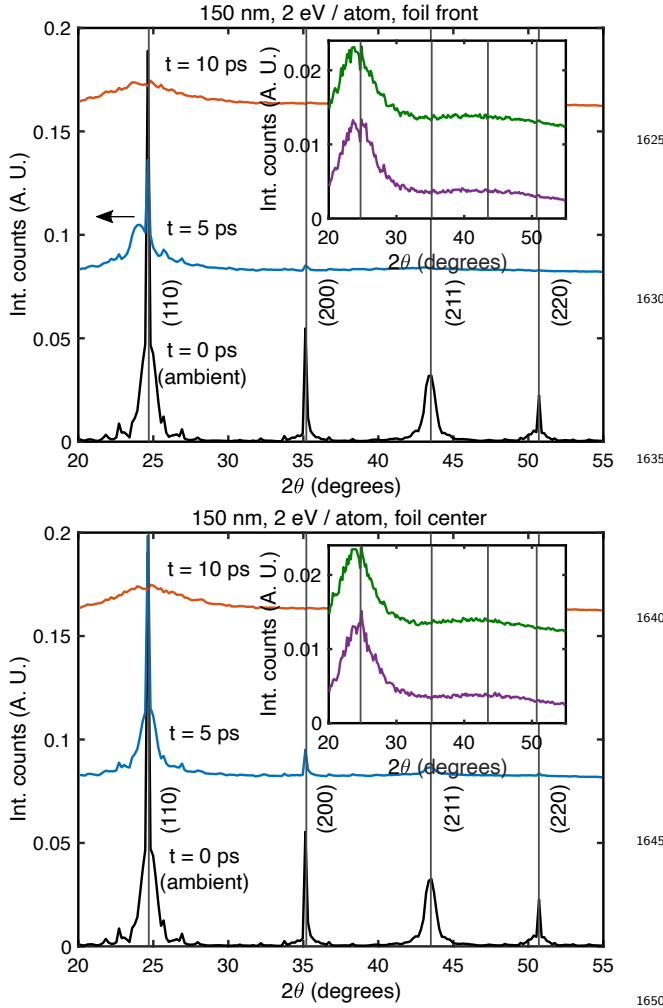


FIG. 15 Synthetic x-ray diffraction patterns from simulations with a 150 nm thick foil, heated to 2 eV / atom. The top panel shows diffraction patterns obtained from a volume of $15 \times 15 \times 15$ unit cells located at the foil front surface, whilst the bottom panel shows diffraction from a region of similar volume, this time located at the center of the foil (which remains under stress confinement conditions for longer). In both panels, the black pattern corresponds to the diffraction pattern from a tantalum crystal at ambient conditions (after equilibration to 300 K), whilst the blue and orange patterns are obtained at 5 and 10 ps, respectively. At the front of the foil the (110) Bragg peak is shifted from the ambient position, at a time of 5 ps, due to tension. The insets show the diffraction patterns taken at 30 and 50 ps, with a re-scaled vertical axis to emphasize the broad liquid peaks. The vertical lines indicate the location of the diffraction peaks from the Miller planes labelled with (h,k,l) indices at the bottom of each line. The panels show that the lattice has melted by 30 ps, with the sharp peaks replaced by broad liquid peaks.

radiation, as well as changing the region of interest and the time at which it is probed. The computational cost of the algorithm is reduced by specifying the angular range of interest. In the proceeding analysis, the XRD patterns are obtained for diffraction angles in the range $2\theta = 20$ - 55° , which captures reflections from the tantalum (110), (200), (211) and (220) Miller planes.

Synthetic XRD patterns are presented for the 150 nm thick foil, heated to 2 eV / atom, in Fig. 15. The top

and bottom panels correspond to diffraction data from the upstream free surface and center of the foil, respectively. In each case, the black pattern corresponds to diffraction from tantalum at ambient conditions (taken after the crystal has been equilibrated to 300 K), showing peaks at the angles associated with the labelled Miller planes. The blue and orange lines indicate the XRD patterns at 5 and 10 ps, respectively. The peaks corresponding to the (110), (200) and (211) planes are still present at 5 ps, however these peaks decrease in intensity and become broad and liquid-like at 10 ps. At the front of the foil, at a time of 5 ps, the location of the (110) Bragg peak is shifted from the ambient position due to tension. The insets in each panel show the XRD patterns at 30 and 50 ps (after the cut-off time of the TTM module), with a re-scaled vertical axis to provide a clearer view of the broad, liquid-like peaks associated with molten material. The synthetic XRD profiles compliment the space-time plot of the lattice temperature in Fig. 4 (f), demonstrating that the front and center of the foil undergo melting upon heating to 2 eV / atom.

Convergence testing of the TTM-MD simulations

It is known that the ablation dynamics have a dependence on the size of the simulation cross-section⁶⁸. There are two constraints on the cross-sectional dimensions of the presented simulations. The first is that the cross-section must be greater than the diameter of a critically sized void, to avoid incorrectly reporting the ablation threshold. This condition is indeed met in the simulations, given that the cross-section is 19.8 nm and the size of the critical void is ≈ 4 Å.

As reported in Ref. 68 there is an additional consideration in the case of periodic boundary conditions. If the cross-section is too small then voids can interact with their mirror image across the periodic boundary, resulting in tensile stress with an artificially high magnitude. To check this is not the case in the current study, convergence tests were conducted by running the 300 nm simulations with different transverse dimensions. The cross-sections were 15×15 , 30×30 and 60×60 unit cells, and for each case the value of the hydrostatic pressure at the time of maximum tensile stress was recorded. Note that it was impractical to increase the cross-section beyond 60×60 unit cells (corresponding to 6,544,800 atoms for the 300 nm case) due to the required run time of the simulations.

The results of the convergence testing are presented in Fig. 16. Comparing the 15×15 and 30×30 unit cell simulations, the magnitude of the tensile stress at the ablation threshold is 6.3 % greater in the simulation with the smaller dimensions. In contrast, the magnitude of the stress reported in the 30×30 and 60×60 unit cell simulations at the ablation threshold differ by only 0.2%. The simulations presented in this study, with cross-sectional size of 60×60 unit cells, are therefore sufficiently large to prevent overestimation of the magnitude of tensile stress.

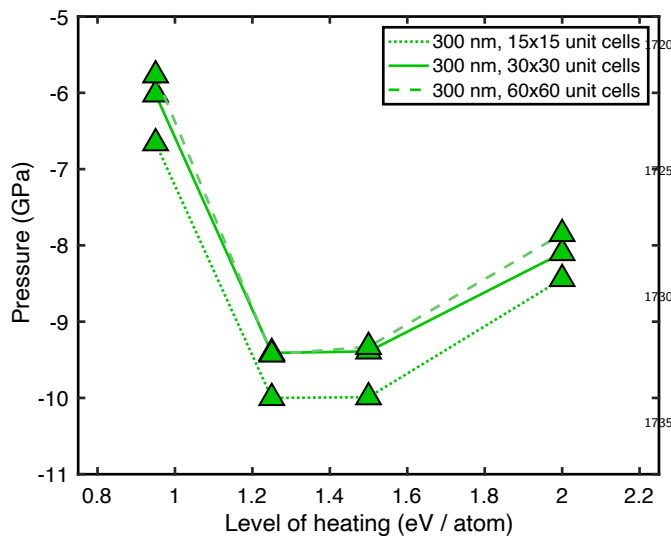


FIG. 16 Pressure at the time of maximum tension, presented as a function of the level of heating for the 300 nm simulations. Different transverse dimensions are tested to determine the simulation size necessary to prevent overestimation of the tensile stress.

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