Supplementary Information (SI) for Digital Discovery.

Supporting Information

Aikaterini Vriza,[1*!] Uma Kornu,[2*] Aditya Koneru,[3] Henry Chan,[1,2,!] Subramanian KRS Sankaranarayanan[1,2,!]

[1] Center for Nanoscale Materials, Argonne National Laboratory, Lemont, IL, 60439, USA

[2] Department of Mechanical and Industrial Engineering, University of Illinois, Chicago, IL, 60607, USA

[3] Argonne Leadership Computing Facility, Argonne National Laboratory, Lemont, IL, 60439, USA

Code repository: https://github.com/ANL-NST/LAMMPS-Agents

Contents

* Equal contribution

! Authors for correspondence – kvriza@gmail.com, hchan@anl.gov; skrssank@anl.gov

1. Agentic system architecture

The three main layers of the system are the agent factory, where we register all the agents, the specialized tools, where several functions for each property calculation and file handling are carefully designed and the function registry where we assign the appropriate functions to each of the agents.

1.1 Agent factory

The main agents developed for this work are described below:

1. **Structure creation agent**: Responsible for creating the appropriate structure file using the Atomsk package.[1,2]
2. **LAMMPS input agent**: Responsible for the generation of the LAMMPS input file tailored to the property prediction requested from the user.
3. **Potential agent**: Responsible for identifying the correct potential file either from the appropriate GitHub links or from performing google search in collaboration with the Websurfer agent.
4. **Melting point calculation agent**: Responsible for handling calculations for melting point estimation.
5. **Elastic constants agent**: Responsible for handling calculations for elastic constants calculation.
6. **Phonopy agent**: Responsible for handling calculations for phonon dispersion calculation.
7. **Results analysis agent – with integrated vision:** Responsible for analysis the results of the simulations by checking the output files. The agent is integrated with vision functions that can assess OVITO frames.
8. **Websurfer Agent**: Tasked to perform google searches and access relevant LAMMPS documentation.
9. **LAMMPS input reviewer agent**: Responsible to review the LAMMPS input files when an execution error appears and provide feedback and corrections.
10. **HPC agent**: Responsible to submit jobs on an HPC cluster (Carbon cluster) if available or run the simulations locally.
11. **Administrator with integrated executor**: Coordinates the interactions between the human user and the agents. Executes the functions that are associated with the agents.

Each agent has a detailed system message to describe the functionalities and main workflow that should execute.

1.2 Specialized tools

Several specialized tools were designed for each individual calculation and for implementing the different packages like ASE, OVITO, Phonopy (phonopy-2.40). Also to connect to HPC supercomputer (Carbon) the user establish connection and set up their username and password.

1.3 Function registry

The function registry is assigning a function to an agent and via connection to the administrator which is connected to the executor.

```python
# Validation functions for the potential agent
validation_functions = [
    (validate_potential_file_wrapper, "validate_potential_file",
     "Validate potential file format and parameters. Parameters: filepath (str), element (str)"),
    (download_from_url, "download_from_url", "Download file from URL. "
    "Parameters: url (str), filename (str)"),
    (verify_download_success, "verify_download_success",
     "Verify downloaded file is real (not dummy). Parameters: filepath (str), expected_element (str)"),
]

for func, name, description in validation_functions:
    register_function(
        func,
        caller=self.potential_agent,
        executor=self.lammps_admin,
        name=name,
        description=description,
```

Supplementary Fig. 1. Example of registering functions to the potential agent for downloading a potential file from a URL and verifying it is not empty or wrong.

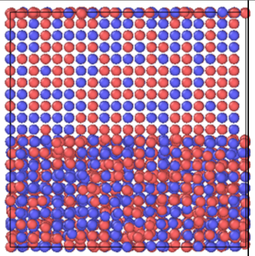
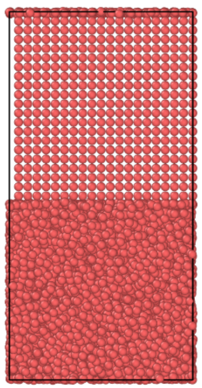
1.4 Vision models

To evaluate the performance of the vision agents and their capabilities to provide a meaningful description of the image frames generated from the ResultsAnalysis agent and the OVITO visualization, we created a small image benchmark (Ovito frames benchmark). The main goals are to be able to identify which vision model is more capable in 1) deciding in a structure corresponds to a 50:50 liquid-solid interface. If the interface is not 50:50 the LAMMPS input creator should update the LAMMPS input file and unfreeze more atoms and resubmit the simulation until a 50:50 solid liquid interface is observed which will be further used as a starting structure for the melting point calculation and 2) deciding whether a structure is fully melted or not. If the structure is not fully melted, then the agents should resubmit a simulation by modifying several conditions until full melting is achieved. These decisions are important for the correct and complete execution of a melting point calculation. We used 6 diverse OVITO frames for performing the evaluation of the vision models for each of the two tasks. We run each query for three times and accessed the reproducibility of the response. The two system prompts used for the vision agents for each task are provided below:

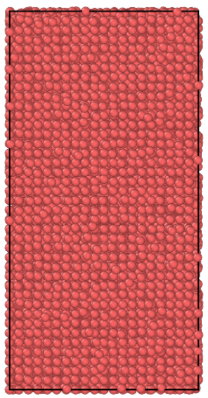
```
"""
analyze_solid_liquid_interface – system prompt

You are a Vision Agent specialized in analyzing molecular dynamics simulation
visualizations created by OVITO.
Check if there are two phases shown in the image, such as a solid-liquid
interface.
We need to have a structure that is around 50:50 solid-liquid. So around 50%
of the structure should look disordered and liquid-like, while the other 50%
should look crystalline and solid-like.
If it is not 50:50 then we need to resubmit the LAMMPS simulation and freeze
or unfreeze atoms.
"""


"""
```

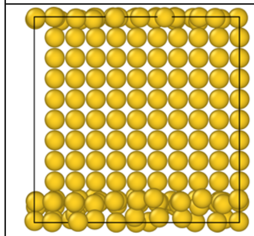
```
analyze_melting_point_simulation – system prompt

You are a Vision Agent specialized in analyzing molecular dynamics simulation
visualizations created by OVITO.

For a melting point simulation, please decide if the structure is fully melted
or not.
For a fully melted structure, look for:
- Random distribution: The atomic positions appear to be randomly distributed
throughout the entire image. There is no discernible long-range order or
pattern in the arrangement of atoms.
- Absence of lattice structures: There are no visible lattice structures or
periodic patterns that would indicate a crystalline arrangement. The atoms are
positioned in a disordered manner across the entire image.
- Non-aligned packing: The atoms are closely packed but not aligned in any
regular manner. This is characteristic of a liquid-like structure where atoms
can move freely.
- Disordered, fluid-like appearance: The overall arrangement looks disordered
and fluid-like, consistent with a fully melted state. There is a uniform
randomness to the atomic positions across the entire image.
- No ordered regions: There are no visible regions of ordered clusters or rows
of atoms that would suggest partial crystallinity. The disorder is consistent
throughout the image.
- ALWAYS ignore colour coding

If the structure is not fully melted, recommend parameter adjustments:
- Increase temperature range
- Extend simulation time (increase timesteps)
- Adjust heating rate (slower heating)
- Check potential parameters

Review your assessment carefully before concluding and providing an answer.
"""
```
Supplementary Fig. 2. System prompt for the visual analysis performed from the vision agent.

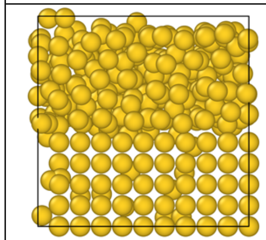
Supplementary Table 1. Comparison of image reasoning accuracy between o3, GPT-4o and QWEN3-VL-8B-Thinking models on the OVITO benchmark for deciding if a frame shows a 50:50 solid-liquid interface or not. The incorrect answers are highlighted with red.

OVITO frame	gpt-4o response	o3 response	qwen3-vl-8b-thinking
	Analysis result: The image depicts two distinct phases: a crystalline structure in the upper part and a disordered, liquid-like structure in the lower part. However, it appears that the solid-like region (upper part) makes up slightly more than 50% of the structure, while the liquid-like region (lower part) constitutes less than 50%. This suggests an imbalance in the intended 50:50 distribution. To achieve a more balanced 50:50 solid-liquid interface, you may need to adjust the simulation parameters, potentially by freezing or unfreezing some of the atoms in the system.	Analysis result: • Top region: atoms are on a perfect lattice; this is the crystalline (solid) part. • Bottom region: atoms are clearly disordered; this is the liquid part. • The interface between the two is reasonably sharp and flat. Relative thickness: The picture height is ~160 px. – Crystalline part ≈ 85–90 px (≈ 55–60 %). – Liquid part ≈ 70–75 px (≈ 40–45 %). Conclusion: The system is close to a 50 : 50 distribution, but it is still somewhat solid-heavy (≈ 55–60 % solid vs. ≈ 40–45 % liquid). If you need a stricter 50 : 50 split, unfreeze (or melt) one or two additional crystalline layers, or freeze one or two liquid layers, then re-equilibrate. Otherwise, the current configuration is acceptable for most purposes	Analysis result: To determine if the visualization shows an approximately 50:50 solid-liquid interface, we analyze the **structural order** of the atoms (spheres) in the image: 1. **Solid Phase Identification**: The top portion of the image displays a **highly ordered, crystalline structure**. Atoms (red and blue spheres) form a regular, repeating lattice with consistent spacing and alignment. This is characteristic of a solid phase (e.g., a crystalline solid). 2. **Liquid Phase Identification**: The bottom portion of the image shows **disordered, random arrangement** of atoms. There is no clear repeating lattice, and atom positions appear more fluid and irregular. This matches the typical behavior of a liquid phase. 3. **50:50 Split Check**: Visually, the image is divided nearly equally between the ordered (solid) top half and disordered (liquid) bottom half. This aligns with the requirement for an **approximately 50:50 solid-liquid split** (i.e., roughly half the structure is solid-like, half is liquid-like). ### Conclusion: The visualization shows a **valid solid-liquid interface** with an approximately 50:50 solid-liquid distribution. **No resubmission or simulation adjustments are needed**.
	Analysis result: Based on the image, it appears there is a clear distinction between two phases. The top part seems to show a	Analysis result: Assessment of the snapshot 1. Presence of two phases • Upper portion: atoms occupy	Visual Analysis: Top half: The atoms are arranged in a highly ordered, crystalline lattice — clearly visible as a regular hexagonal or FCC-like pattern. This is characteristic of a solid phase. Bottom half: The atoms appear disordered, with no long-

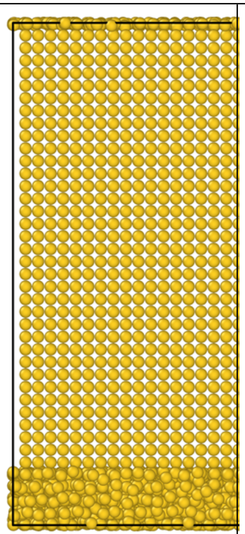
*
! ndence – kvriza@gmail.com, hchan@anl.gov; skrssank@anl.gov

	crystalline, ordered structure representing the solid phase, while the bottom part looks disordered, indicating a liquid-like phase. Visually, the solid and liquid phases take up approximately equal space, suggesting the structure is around 50:50 solid-liquid. This implies that the setup meets the desired balance. No need for adjustments or resubmission at this point.	well-defined lattice sites → crystalline / solid. • Lower portion: atoms are randomly packed without long-range order → liquid-like. → Two distinct phases are clearly present, separated by a fairly sharp horizontal interface. 2. Phase fraction (solid : liquid) Visually, the interface cuts the column almost in the middle. • Solid ~45–50 % of the cell height • Liquid ~50–55 % of the cell height 3. Verdict This is close enough to the desired 50 : 50 ratio. No resubmission or additional freezing/unfreezing of atoms is necessary at this stage.	range order and irregular packing — typical of a liquid phase. There is a distinct horizontal boundary between the two regions, indicating a well-defined solid-liquid interface. Quantitative Estimate: The system appears to be divided approximately equally into top (solid) and bottom (liquid) halves. The interface is sharp and clean, suggesting deliberate setup (e.g., via freezing/unfreezing atoms or temperature gradient). Visually, the ratio is very close to 50:50, possibly slightly more solid than liquid, but within acceptable tolerance. Conclusion: Yes, there are two phases shown: a solid phase (top) and a liquid phase (bottom). The structure is approximately 50:50 solid-liquid, meeting the required condition. Recommendation: Since the visual inspection confirms a near-perfect 50:50 split, no resubmission of the LAMMPS simulation is needed.
	Analysis result: In the image provided, the structure appears almost entirely uniform, suggesting that it is predominantly in a single phase. To establish a 50:50 solid-liquid interface, the structure should exhibit clear regions of both crystalline (orderly) and disordered (liquid-like) phases. This image does not show a distinct demarcation between solid	Analysis result: Assessment • The entire simulation box is filled with atoms arranged on a perfect, periodic lattice. • No visibly disordered, diffuse, or less-dense region can be seen. • Therefore, the configuration is essentially 100 % crystalline (solid) and 0 % liquid. Conclusion The current snapshot is not close to a 50:50 solid-liquid mixture. You will need to re-run the LAMMPS preparation step and either:	Analysis result: To determine if the image shows a 50:50 solid-liquid interface, we analyze the structural characteristics: 1. **Observation of the structure**: The image displays a **uniform, highly ordered lattice of red spheres** with no visible regions of disorder (typical of a liquid phase). The atoms are arranged in a regular, crystalline pattern throughout the entire visualization. 2. **Comparison to the 50:50 requirement**: A 50:50 solid-liquid interface would require **distinct regions**—one half with crystalline order (solid-like) and the other with disordered, random atomic positions (liquid-like). However, this image shows **no evidence of disorder**;

	and liquid phases, so you may need to adjust your LAMMPS simulation to achieve the desired configuration.	1. Melt (unfreeze / heat) roughly half of the box, or 2. Freeze half of a fully molten configuration, to create a stable solid-liquid interface. Only after you see a clear disorder/crystalline contrast occupying roughly equal volumes should you proceed to production runs.	the entire structure appears fully solid-like (crystalline). There is no transition or mixed phase visible. 3. **Conclusion**: The structure does **not** meet the 50:50 solid-liquid ratio requirement. The entire visualization is dominated by a solid-phase structure, with no liquid-like regions. **Recommendation**: Resubmit the LAMMPS simulation to generate a proper solid-liquid interface (e.g., by selectively freezing/unfreezing atoms to create a 50:50 phase separation).
	Analysis result: The image shows a structure with a crystalline and ordered appearance, indicative of a solid phase. However, there is a section at the bottom that displays some disorder, suggestive of a liquid phase. From this visualization, it seems that the structure is predominantly solid with a smaller portion being liquid-like. It does not appear to be a 50:50 solid-liquid distribution. You may need to adjust your LAMMPS simulation, possibly modifying the temperature or using different parameters to achieve a more balanced solid-liquid interface.	Analysis result: Assessment • The entire simulation box is filled with atoms arranged on a perfect, periodic lattice. • No visibly disordered, diffuse, or less-dense region can be seen. • Therefore, the configuration is essentially 100 % crystalline (solid) and 0 % liquid. Conclusion The current snapshot is not close to a 50:50 solid-liquid mixture. You will need to re-run the LAMMPS preparation step and either: 1. Melt (unfreeze / heat) roughly half of the box, or 2. Freeze half of a fully molten configuration, to create a stable solid-liquid interface. Only after you see a clear disorder/crystalline contrast & C:/Users/kvriz/miniconda3/envs/autogen/python.exe c:/Users/kvriz/Desktop/LAMMPS-Agents/src/tools/specialized_tools/vis	To determine if the image shows a **50:50 solid-liquid interface**, we analyze the structural arrangement of atoms: 1. **Solid-like regions**: Crystalline structures exhibit *ordered, repeating lattice patterns*. In the image, all yellow spheres form a **perfectly regular grid** (rows and columns are uniformly aligned). This indicates a fully crystalline structure with **no disorder**. 2. **Liquid-like regions**: Liquid structures have *disordered, randomly arranged atoms*. The image contains **zero regions of disorder**—every atom follows the same lattice pattern. ### Key observation: The entire structure is solid-like (100% crystalline), with **no liquid-like (disordered) portion**. For a 50:50 split, we would need ~50% of the structure to appear disordered (liquid-like) and ~50% ordered (solid-like). This image fails to meet this requirement. ### Conclusion: The visualization does **not** show a 50:50 solid-liquid interface. The simulation must be resubmitted with adjustments to atom freezing/unfreezing to create a balanced solid-liquid interface.

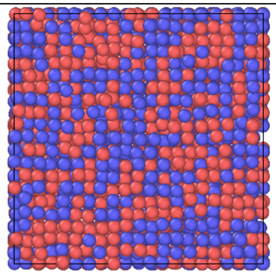
		ion_manager.pyest> Analysis result: The configuration is almost entirely crystalline. The atoms are arranged on a well-defined lattice throughout the cell; only a very thin, slightly rumpled layer at the very bottom shows any noticeable disorder. That disordered region occupies far less than 50 % of the volume—more like a few percent. Therefore, this snapshot is not close to a 50:50 solid–liquid mixture. You will need to adjust the simulation (e.g., melt more of the system or unfreeze additional atoms) and re-equilibrate to obtain a roughly half-solid, half-liquid interface.	
	Analysis result: The image shows a clear distinction between a solid and a liquid phase. The top half appears disordered, indicative of a liquid-like structure, while the bottom half looks crystalline, suggesting a solid-like structure. However, it seems that the solid and liquid phases are not exactly 50:50; the liquid phase appears to occupy a slightly larger portion than the solid phase. You may want to adjust the LAMMPS simulation to achieve a more balanced 50:50 solid-liquid	Analysis result: From the visualization you supplied, the system does appear to contain two clearly distinguishable phases: • Lower half – atoms are arranged on a periodic lattice with straight, well-aligned atomic rows: this is the crystalline (solid) region. • Upper half – atomic positions lack long-range order and the packing looks noticeably more random: this is the disordered (liquid-like) region. The interface is roughly at the mid-height of the simulation cell, so the solid and liquid portions each occupy close to one-half of the volume. That satisfies the ≈50 : 50 requirement for a balanced solid–liquid coexistence run. No resubmission or additional	Analysis result: To analyze the provided visualization for a **50:50 solid-liquid interface** (as requested), here's the step-by-step assessment: ### 1. **Identify Key Structural Features** - **Solid-like (crystalline) regions**: Characterized by *ordered, repeating patterns* (e.g., regular lattice, fixed atomic positions). - **Liquid-like (disordered) regions**: Characterized by *random, irregular arrangements* (e.g., no long-range order, fluctuating positions). ### 2. **Examine the Image** The visualization shows **yellow spheres arranged in a uniform, grid-like lattice** across the entire frame. There are no visible regions with disordered atomic positions, irregular spacing, or signs of a liquid phase. The structure is **completely ordered** (consistent spacing, periodic alignment) and lacks the chaotic, non-repeating arrangement typical of a liquid.

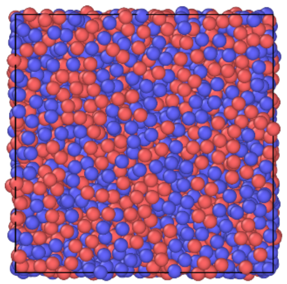
	interface. Consider freezing some atoms in the liquid-like region or promoting disorder in the solid-like region to achieve this.	freezing/unfreezing of atoms looks necessary at this stage.	### 3. **Check for 50:50 Solid-Liquid Balance** For a 50:50 solid-liquid interface, **half the structure must look disordered (liquid-like) and the other half crystalline (solid-like)**. In this image: - **No liquid-like regions** are present (no disordered areas). - The entire structure is solid-like (ordered crystalline lattice). - The ratio is **100% solid : 0% liquid**, which fails the 50:50 requirement. ### 4. **Conclusion** The image does **not** show a 50:50 solid-liquid interface. Instead, it depicts a *pure crystalline structure* (no liquid phase). To meet the 50:50 requirement, the LAMMPS simulation must be adjusted (e.g., by freezing/unfreezing atoms, modifying temperature, or adjusting boundary conditions) to create a distinct solid-liquid interface with approximately equal areas of order and disorder. ### Recommendation Resubmit the LAMMPS simulation with modifications to induce a 50:50 solid-liquid interface (e.g., partial freezing of atoms to create a solid region, while keeping other atoms in a liquid state). **Status**: ✕ Does **not** meet the 50:50 solid-liquid interface criterion. Resubmit simulation.

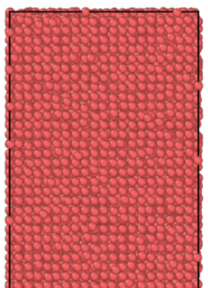
	Analysis result: The image shows a structure with a mostly crystalline, solid-like arrangement. There is a small region near the bottom that appears more disordered, indicating a possible liquid-like state. However, it does not seem to be approximately 50:50 solid-liquid. The solid region is dominant. You may need to adjust the simulation by altering the freezing or unfreezing of atoms for a better balance.	Analysis result: The snapshot shows only a small disordered "cap" along the bottom edge—perhaps 10–15 % of the cell height—while the remaining ≈ 85–90 % of the atoms are still in a perfectly ordered crystal lattice. That means the present configuration is almost entirely solid, not the desired ≈ 50 : 50 solid-liquid mixture. Recommendation • Rerun / continue the LAMMPS run and melt a larger portion of the sample, or un-freeze a thicker section of atoms, until roughly half of the slab has lost its lattice order. • Alternatively, keep the melted part as is but freeze an equal thickness of atoms at the opposite end, then heat only the unfrozen middle so that half becomes liquid. • Once the disordered region occupies about half of the simulation box, re-export the frame for further analysis. In short: the current frame is not suitable; you will need to adjust the melt/freeze regions and resubmit the simulation.	Analysis result: To determine if the simulation visualization shows a **50:50 solid-liquid interface**, we analyze the structural features in the provided image: ### Key Observations: 1. **Solid Phase Characteristic**: In a crystalline (solid) phase, atoms form a **regular, repeating lattice** (e.g., uniform spacing, predictable geometry). 2. **Liquid Phase Characteristic**: In a liquid phase, atoms exhibit **disorder** (e.g., irregular spacing, no long-range order). 3. **50:50 Requirement**: The image must show **~50% of the structure as disordered (liquid-like)** and **~50% as ordered (solid-like)**. ### Analysis of the Image: - The visualization shows a **uniform grid of yellow dots** throughout the entire frame. - There is **no visible disordered region** (no irregular spacing, no loss of long-range order) to indicate a liquid phase. - The structure appears to be **fully crystalline (solid)**—consistent with a single-phase solid structure, not a 50:50 solid-liquid interface. ### Conclusion: The image does **not** show a 50:50 solid-liquid interface. The entire structure is uniform and ordered, indicating a pure solid phase with **no liquid-like region**. ### Recommendation: Resubmit the LAMMPS simulation to: - Adjust boundary conditions or temperature to induce melting (to create a liquid phase). - Ensure the simulation setup (e.g., freezing/unfreezing

			atoms) produces a clear solid-liquid interface with **~50% liquid and ~50% solid**. No further visualization is needed—this image does not satisfy the required phase separation.

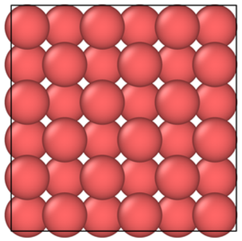
Supplementary Table 2. Comparison of image reasoning accuracy between o3, GPT-4o and QWEN3-VL-8B-Thinking models on the OVITO benchmark for classifying a structure as fully melted or not melted yet. The incorrect answers are highlighted with red. We observed that 4o model was confused in some cased with alloys due to the different coloring, but o3 did not have this issue.

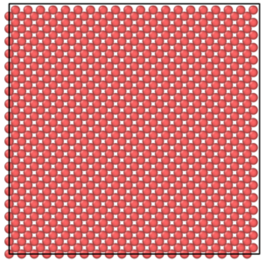
OVITO frame	gpt-4o response	o3 response	qwen3-vl-8b-thinking
	Analysis result: The structure is **fully melted**. Reasons: • The atoms are distributed randomly across the entire simulation box; no long-range periodicity or straight rows are visible. • There are no discernible lattice planes or repeating	Analysis result: Assessment: **NOT fully melted.** Reasoning • The atoms form recognizable straight rows/columns along the box edges, revealing a residual lattice. • Inter-atomic spacing is almost identical	Analysis result: To determine if the structure is fully melted, we analyze the visualization against the criteria for a **fully melted state**: ### Step 1: Assess Structural Order - **Random distribution & absence of lattice structures**: The image shows a **clear, periodic grid-like arrangement** of atoms (red and blue spheres form a regular, repeating pattern across the entire image). There is *long-range order* (e.g., rows and columns of atoms are uniformly spaced and aligned), which is **not** characteristic of a melted structure. In a fully melted state, atomic positions should be *randomly distributed* with no discernible repeating pattern. - **Non-aligned packing & fluid-like appearance**: The atoms are *highly aligned* in a grid, not freely packed or fluid-like. A melted structure lacks this

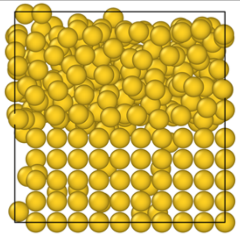
	patterns that would indicate residual crystalline order. • Packing is dense yet non-aligned, giving the image a uniform, fluid-like appearance. • No localized regions show crystalline clusters—the disorder is consistent throughout. No further parameter adjustments are necessary.	throughout, suggesting long-range periodicity rather than the random, fluid-like packing of a liquid. • Patches of red and blue atoms appear in ordered domains rather than being uniformly scrambled. Recommended adjustments 1. Increase the target temperature (or run a short additional ramp) to supply enough thermal energy. 2. Extend the simulation (more timesteps) so atoms have time to lose their lattice memory. 3. Use a slower heating rate; rapid ramps can leave kinetically trapped crystalline remnants. 4. Re-check interatomic potential parameters to ensure they accurately reproduce melting behaviour for this system.	rigid, ordered arrangement. - **No ordered regions**: The entire image exhibits **uniform, long-range order** (e.g., every row/column has identical spacing), meaning there are no regions of disorder. In a melted state, disorder should be consistent across the entire simulation. ### Step 2: Conclusion on Melted State This structure is **not fully melted**. Instead, it shows **crystalline order** (evidenced by the periodic lattice structure). The absence of random atomic distribution and presence of long-range order confirm it remains in a solid-like state. ### Step 3: Recommended Parameter Adjustments To achieve a fully melted structure, adjust the simulation parameters to reach the **melting transition**: 1. **Increase temperature range**: Ensure the simulation temperature exceeds the system's melting point (e.g., raise the final temperature above the expected melting temperature). 2. **Extend simulation time**: Allow sufficient time for thermal equilibration and phase transition (e.g., increase the number of timesteps to observe melting dynamics). 3. **Adjust heating rate (slower)**: A slower heating rate may allow the system to gradually absorb energy, facilitating a more accurate melting transition (rapid heating could trap the system in a solid state). 4. **Check potential parameters**: Verify that the interatomic potential correctly models the material's melting behavior (e.g., ensure the potential is appropriate for the system's intermolecular forces). ### Final Answer The structure is **not fully melted**—it exhibits crystalline order (periodic lattice structure). To observe a fully melted state, **increase the temperature range**, **extend simulation time**, and/or **adjust the heating rate to be slower**.
	Analysis result: Assessment: **Fully melted** Reasoning • The atoms fill the entire simulation cell with a uniform, random-looking distribution. • No straight rows, planes, or repeating patterns characteristic of a	Analysis result: Assessment: **The structure is fully melted.** Reasoning: • Atomic positions are randomly distributed throughout the simulation box; no straight rows, planes, or periodic patterns characteristic of a	Analysis result: ### Analysis of Molecular Dynamics Simulation Visualization **Assessment: The structure is fully melted.** #### Reasoning: To determine if the structure is fully melted, we evaluate the key criteria for a liquid state (ignoring color coding, as specified): 1. **Random Distribution**: The atomic positions in the image lack any discernible long-range order. There

	crystalline lattice are visible. • Packing is dense but not aligned; there is no indication of partially ordered clusters or domains. • Disorder is consistent throughout the whole box—no regions retain long-range order. Therefore, the structure can be considered fully melted; no additional parameter adjustments are required.	crystal lattice are visible. • The packing is dense but non-aligned, giving a disordered, fluid-like appearance over the entire image. • There are no isolated regions of ordered clusters; the disorder is uniform from edge to edge. Because the system already exhibits the hallmarks of a liquid, no further parameter adjustments are necessary for complete melting.	are no repeating patterns, grids, or systematic alignments of atoms across the entire visualization. Instead, the atoms appear randomly interspersed throughout the box, consistent with the free, disordered motion of particles in a liquid. 2. **Absence of Lattice Structures**: No periodic or crystalline lattice (e.g., square, hexagonal, or cubic patterns) is visible. In a solid state, atoms would align in a regular, repeating arrangement, but this image shows no such structure. The absence of long-range periodicity confirms the lack of crystalline order. 3. **Non-Aligned Packing**: While atoms are closely packed (as expected in a dense liquid), they are not arranged in any regular, directional pattern. This “random close packing” is characteristic of a liquid, where atoms lack the fixed positional relationships seen in solids. 4. **Disordered, Fluid-Like Appearance**: The overall arrangement appears fluid and homogeneous. There is no evidence of distinct regions with ordered atomic clusters (e.g., rows, columns, or crystalline domains). The uniform randomness of atomic positions aligns with the behavior of a fully melted system, where thermal energy dominates over intermolecular forces, leading to constant atomic motion. 5. **No Ordered Regions**: Upon close inspection, there are no localized regions of order (e.g., small crystalline patches or aligned rows) that might suggest partial crystallinity. The disorder is consistent throughout the entire image, which is consistent with a fully melted system. #### Conclusion: The visualization exhibits all hallmarks of **a fully melted** structure: random atomic distribution, absence of long-range lattice order, and a fluid-like, disordered arrangement. There are no indications of residual crystalline order (e.g., partial melting or solid-like features), so **the structure is fully melted**. *(Note: If the simulation were not fully melted, we would recommend adjusting parameters such as temperature range, simulation time, or heating rate. However, based on this visual analysis, no parameter adjustments are necessary.)*
	Analysis result: The structure is **NOT fully melted.** Observed features:	Analysis result: The structure is **NOT fully melted**. Assessment:	Analysis result: To determine if the structure is fully melted, we analyze the atomic arrangement against the criteria for a fully melted state: ### Key Observations from the Visualization:

	• Clear, periodic lattice arrangement; atoms sit in well-defined rows and planes. • Long-range order across the entire image; no random or fluid-like distribution. • Overall appearance is that of an intact crystal, not a liquid. Recommended adjustments to reach a melted state: 1. Increase the target temperature (raise the upper limit of the heating schedule). 2. Extend the simulation time / number of timesteps to allow the system to equilibrate at high temperature. 3. Use a slower heating rate so the system can overcome barriers and properly disorder. 4. Double-check the interatomic potential and thermostat settings to be sure they allow melting at the expected temperature. Apply one or a combination of these changes and re-run the simulation until the lattice order disappears and the atoms adopt a fully random, fluid-like distribution.	• Clear long-range order and periodic packing are visible throughout the sample. • Atoms form well-aligned rows/planes that indicate a crystalline lattice. • No regions of uniform, random, fluid-like disorder are present. Recommendations to reach a fully melted state: 1. Increase the target temperature (or temperature range) beyond the current value. 2. Extend the simulation time so the system can equilibrate at high temperature (more timesteps). 3. Use a slower, more gradual heating rate to avoid kinetic trapping and ensure uniform melting. 4. Verify/adjust the interatomic potential parameters to ensure they reproduce realistic melting behavior for this material.	- **Long-range order is evident**: The atoms are arranged in a **clear, periodic grid pattern** (rows and columns of atoms aligned uniformly). This is characteristic of a *crystalline solid*, not a disordered liquid. - **No random distribution**: The atoms are not randomly positioned; instead, they follow a highly regular lattice structure across the entire image. - **Lattice structures are present**: The image shows explicit periodicity (e.g., repeating rows/columns of atoms), which is absent in a fully melted state. - **No fluid-like appearance**: The structure lacks the uniform randomness and disordered packing typical of liquids. Instead, it has rigid, aligned atomic positions. - **Ordered regions dominate**: The entire image exhibits consistent order, with no regions of partial crystallinity or disorder—this is the hallmark of a solid, not a melted state. ### Conclusion: The structure is **not fully melted**. The presence of a periodic grid pattern indicates the sample remains in a **crystalline solid state**. Melting would destroy this long-range order, resulting in random atomic positions and a fluid-like appearance. ### Recommended Parameter Adjustments: To achieve a fully melted state in future simulations, consider: 1. **Increase temperature range**: Ensure the final temperature exceeds the predicted melting point. 2. **Extend simulation time**: Allow sufficient timesteps for thermal equilibration and melting to occur (a short simulation may not capture the transition). 3. **Adjust heating rate (slower heating)**: Rapid heating may skip the melting transition; a slower ramp allows the system to relax into a melted state. 4. **Check potential parameters**: Verify the interatomic potential correctly models melting (e.g., validate with prior literature for the material/system). *Note: Always ignore color coding (as instructed), as color is irrelevant to structural analysis of atomic arrangements.*

	Analysis result: Assessment: **NOT fully melted** Reasoning: • Clear long-range order – atoms are arranged in straight rows and columns. • Obvious lattice/periodic packing – regular square grid with identical spacing. • No sign of the random, fluid-like distribution characteristic of a liquid. Recommendations to reach a fully melted state: 1. Increase temperature (raise the upper bound of the heating schedule). 2. Extend the simulation time so atoms can overcome energy barriers (more timesteps). 3. Apply a slower heating rate to avoid superheating and give the structure time to disorder. 4. Verify/adjust interatomic potential parameters to be sure they reproduce realistic melting behaviour.	Analysis result: The structure is **NOT fully melted.** The atoms are arranged in a perfectly ordered, periodic lattice with clear alignment and long-range order—typical of a crystalline solid. Recommended adjustments to reach a fully melted state: 1. Increase the target temperature or widen the temperature range. 2. Extend the simulation time (run for more timesteps) to let melting progress. 3. Use a slower, more gradual heating rate to avoid superheating artifacts. 4. Re-examine and, if necessary, refine the interatomic potential parameters to ensure they reproduce the correct melting behavior.	Analysis result: The structure is **not fully melted**. ### Reasoning: 1. **Long - range order and lattice structure**: The atoms are arranged in a highly regular, periodic grid (square lattice). This is characteristic of a **crystalline solid**, where atoms occupy fixed positions in a repeating pattern. There is no randomness in the atomic arrangement—every row and column aligns perfectly, indicating long - range order. 2. **Absence of liquid - like features**: For a fully melted structure, atoms should show random distribution with no discernible patterns or aligned rows/columns. In this image, the uniform spacing and alignment of atoms contradict the “disordered, fluid - like” appearance required for a liquid phase. 3. **No disorder**: The image lacks the “random distribution throughout the entire image” and “non - aligned packing” described as hallmarks of a liquid state. Instead, it exhibits rigid, ordered clustering. ### Recommended Parameter Adjustments (to achieve melting): - **Increase temperature** above the estimated melting point (if the current temperature is below this threshold). - **Extend simulation time** (increase timesteps) to allow sufficient thermal energy to disrupt the lattice and induce melting. - **Verify potential parameters** (e.g., force field accuracy) to ensure proper repulsive/attractive interactions that mimic real - world melting behavior. This assessment ignores color coding (as instructed) and focuses solely on atomic positional order, confirming the structure remains solid.

	Analysis result: The structure is **NOT fully melted**. Assessment • Clear, long-range, periodic lattice pattern is visible. • Atoms are arranged in perfectly aligned rows and columns with no signs of disorder. • There are no randomly distributed or fluid-like regions. Recommendations to achieve melting 1. Increase the target temperature or widen the temperature ramp. 2. Extend the simulation time (more timesteps) to allow the system to equilibrate at high temperature. 3. Apply a slower, more gradual heating rate so the lattice can break down uniformly. 4. Re-check interatomic potential and thermostat parameters to be sure they allow melting to occur. Implementing one or a combination of these adjustments should help the crystal fully transition to a liquid state.	Analysis result: Assessment: **NOT fully melted.** Reasoning: • The atoms form a perfectly periodic, grid-like lattice covering the whole snapshot. • Long-range order and straight rows/columns are clearly visible; no random, fluid-like distribution. • The structure therefore remains crystalline. Recommended adjustments to reach a fully melted state: 1. Increase the maximum temperature (raise the target temperature above the current value). 2. Extend the simulation time (run for more timesteps) to allow complete melting. 3. Apply a slower heating rate so atoms can irreversibly leave their lattice sites. 4. Re-examine interatomic potential parameters to ensure they reproduce the correct melting point.	Analysis result: ### Analysis of the Molecular Dynamics Simulation Visualization #### Step 1: Assess for "Fully Melted" Criteria The provided visualization shows a **highly ordered, periodic lattice structure** (atoms arranged in a regular grid). Key observations: - **Long-range order**: The atomic positions exhibit a clear, repeating grid pattern (e.g., uniform spacing between atoms, consistent rows/columns). - **Lattice structures**: The image contains distinct periodicity and alignment, which is characteristic of a **crystalline solid**, not a melted state. - **Absence of disorder**: There is no randomness in atomic positions; the structure is highly symmetric and structured. These features directly contradict the criteria for a *fully melted structure* (e.g., "random distribution," "absence of lattice structures," "non-aligned packing"). Instead, the arrangement is consistent with a **solid phase** (likely crystalline) or a system that has not yet melted. #### Step 2: Reason for Not Being Fully Melted The visualization lacks the disordered, fluid-like appearance required for a melted state. In a fully melted system, atoms would exhibit **local disorder** (no long-range periodicity) and **dynamic motion** (e.g., no fixed positions). Here, the strict grid implies the system remains in a crystalline state. #### Step 3: Recommended Parameter Adjustments To achieve a fully melted structure, the following adjustments are advised: 1. **Extend simulation time (increase timesteps)**: The current simulation may be too short to allow the system to reach equilibrium at the melting point. Longer simulation time enables thermal fluctuations to disrupt the lattice and transition to a liquid state. 2. **Increase temperature range**: If the temperature is below the melting point, the system will not melt. Ensure the simulation runs at or above the expected melting temperature (e.g., by raising the temperature incrementally). 3. **Adjust heating rate (slower heating)**: Rapid heating may cause the system to "skip" over the melting transition or not equilibrate properly. A slower heating rate allows the system to gradually absorb heat and transition from solid to liquid. 4. **Check potential parameters**: If the force field or potential is inappropriate for the material, it may not accurately model melting (e.g., over-stabilized crystal structures). Validate the potential against known properties of the system. ### Conclusion

			The structure is **not fully melted** (it remains in a crystalline or solid-like state). To achieve a fully melted configuration, prioritize extending simulation time, ensuring the temperature exceeds the melting point, and adjusting heating kinetics. **Final Answer**: The structure is **not fully melted**. The visualization shows a periodic lattice structure with long-range order, inconsistent with a liquid state. To resolve this, **extend simulation time**, **increase the temperature range** (to exceed the melting point), and **adjust the heating rate to be slower** to allow equilibration. Additionally, verify potential parameters for accuracy.
	Analysis result: The structure is **NOT fully melted**. Assessment: • Upper region – atoms are randomly arranged, indicating local melting. • Lower region – clear, evenly spaced rows of atoms remain, showing long-range crystalline order. Because a sizeable portion of the simulation cell still retains a lattice pattern, the system is only partially melted. Recommended adjustments to reach full melting: 1. Increase the target temperature or widen the temperature range near the expected melting point. 2. Extend the simulation (more timesteps) to allow the remaining crystalline region to transform. 3. Use a slower heating	Analysis result: Assessment: **NOT fully melted** Evidence from the image • The bottom ~40 % of the box still shows clear, parallel rows of atoms – a crystalline lattice. • The upper region is disordered, but the coexistence of ordered and disordered parts indicates only partial melting. • Long-range order is therefore present in a significant portion of the sample, so the structure is not yet fully liquid. Recommended parameter adjustments 1. Increase the target temperature or widen the temperature ramp to exceed the melting point. 2. Extend the simulation time (more timesteps) at the high-temperature plateau to allow complete melting.	Analysis result: To determine if the structure is fully melted, we analyze the image against the criteria for a fully melted state (random distribution, absence of lattice structures, no ordered regions, etc.). ### Step-by-Step Reasoning: 1. **Lattice Structure Observation**: The lower portion of the image shows a clear, **periodic grid-like arrangement** of atoms. This is characterized by rows of atoms aligned horizontally and vertically, forming a crystalline lattice. Such ordered patterns are inconsistent with a fully melted state (which requires *no* long-range order). 2. **Presence of Ordered Regions**: The bottom section of the visualization exhibits *ordered clusters* (rows of aligned atoms), which directly contradicts the requirement for “no ordered regions” in a fully melted structure. While the top portion may appear more disordered, the presence of any crystalline ordering (even partial) means the system is **not fully melted**. 3. **Disorder vs. Order**: A fully melted structure must display *uniform randomness* across the entire image. Here, the lower half’s lattice structure introduces localized order, violating the criterion for a liquid-like state. ### Conclusion: The structure is **not fully melted**. The presence of a crystalline lattice (ordered rows of atoms) in the lower portion of the visualization indicates residual solid-state order, which is inconsistent with a fully melted state. ### Recommended Parameter Adjustments: To achieve a fully melted state, consider the following: - **Increase temperature range**: Ensure the simulation temperature exceeds the

	rate so the temperature can equilibrate uniformly. 4. Verify/adjust the interatomic potential parameters to ensure they reproduce the correct melting behavior.	3. Use a slower heating rate so that heat diffuses uniformly and prevents a solid core from persisting. 4. Double-check interatomic potential parameters and thermostat/barostat settings to be sure the simulated melting point is accurate. Apply these changes and re-run until the entire volume shows a uniformly random, fluid-like arrangement with no remaining lattice planes.	melting point of the material to disrupt the lattice. - **Extend simulation time**: Allow sufficient timesteps for thermal energy to fully randomize atomic positions and eliminate crystalline regions. - **Adjust heating rate (slower heating)**: Rapid heating may trap the system in intermediate states (e.g., partially melted or metastable phases). A slower heating ramp permits complete melting. - **Check potential parameters**: Verify that the interatomic potential accurately models the material's melting behavior (e.g., incorrect repulsive/attractive terms may prevent proper melting). These adjustments will help transition the system toward a uniformly disordered, fluid-like structure.

2. Atomic simulations case studies

2.1 Lattice constants calculations

```
"""
*INSTRUCTIONS FOR LATTICE CONSTANTS & COHESIVE ENERGY:
    1. Always perform two separate relaxation stages:

        **Step 1: Cell (box) relaxation**
        - The simulation must first relax the **simulation cell volume and shape** under zero external pressure.
        - This means allowing the simulation box to change in **size and angles** in response to internal stresses.
        - The relaxation must include all six lattice degrees of freedom: lengths (`a`, `b`, `c`) and angles (`alpha`, `beta`, `gamma`).
        - This can be done by adding the keywords: etotal vol cella cellb cellc cellalpha cellbeta cellgamma to the thermo_style
        - The goal is to minimize the system’s enthalpy by adjusting the simulation box under zero pressure.

        **Step 2: Atomic position relaxation**
        - Once the simulation cell is relaxed, keep the box fixed.
        - Relax only the internal atomic positions to further minimize the total potential energy.

    2. print Cohesive energy per atom
    3. print Lattice constants (`a`, `b`, `c`, `alpha`, `beta`, `gamma`)


⚠   Relaxing the simulation cell MUST be done explicitly . Do not skip or combine it with atomic relaxation. Always separate these two steps.

Collect Results
Extract the lattice constant and corresponding cohesive energy from each LAMMPS run.

Fit and Analyze
Plot energy versus lattice constant and identify the minimum point. This gives the optimized lattice constant.
"""
```

Supplementary Fig. 3. Detailed instructions to the LAMMPS input creator agent regarding the lattice constants and cohesive energy calculations.

* Equal contribution

! Authors for correspondence – kvriza@gmail.com, hchan@anl.gov; skrssank@anl.gov

a) **StructureCreator** - Creates the crystal structure using Atomsk

```
Calculate the lattice constants and cohesive energy for gold using LAMMPS.

--------------------------------------------------------------------------------
Next speaker: StructureCreator

StructureCreator (to chat_manager):

To calculate the lattice constants and cohesive energy for gold using LAMMPS,
we need to follow these steps:

1. **Create the Unit Cell for Gold:**
   - Gold has an FCC crystal structure.
   - The lattice parameter for gold is approximately 4.078 Å.
   - We will create a unit cell for this calculation.

2. **Relax the Structure:**
   - Before calculating any properties, the structure must be relaxed to
ensure it is in its lowest energy state.

3. **Calculate Lattice Constants and Cohesive Energy:**
   - Use LAMMPS to perform these calculations.

Let's start by creating the unit cell for gold. I'll proceed with creating the
structure.

Suggested tool call (call_GoelO6h05iyoeqxmDlffoR9d): create_structure
Arguments:
{"crystal_type":"fcc","lattice_param":4.078,"element":"Au","size":"1 1
1","output_format":"lammps"}
******************************************************************************
***** Response from calling tool (call_GoelO6h05iyoeqxmDlffoR9d) *****
✅ Crystal structure created successfully:
  📄 File: au_structure.lmp (663 bytes)
  🔬 Structure: fcc Au
  📏 Lattice parameter: 4.078 Å
  📦 Size: 1 1 1
  ⚛    Atoms: 4
```

b) **PotentialManager** - Identifies an appropriate potential file

```
Next speaker: PotentialManager

PotentialManager (to chat_manager):
```

```
The unit cell for gold with an FCC structure and a lattice parameter of 4.078
Å has been created successfully. The structure is saved in the file
`au_structure.lmp`.

Next, we need to select an appropriate potential for gold to perform the
relaxation and energy calculations. For gold, an Embedded Atom Method (EAM)
potential is typically used. Let's proceed to find and download a suitable EAM
potential for gold.
***** Suggested tool call (call_S3ZILRfnwavZvbFAFkgdLaBZ): search_openkim
*****
Arguments:
{"element":"Au","potential_type":"EAM"}

Next speaker: admin

Replying as admin. Provide feedback to chat_manager. Press enter to skip and
use auto-reply, or type 'exit' to end the conversation:

>>>>>>>> EXECUTING FUNCTION search_openkim...
Call ID: call_S3ZILRfnwavZvbFAFkgdLaBZ
Input arguments: {'element': 'Au', 'potential_type': 'EAM'}
admin (to chat_manager):

***** Response from calling tool (call_S3ZILRfnwavZvbFAFkgdLaBZ) *****
🔬 SEARCHING OPENKIM DATABASE FOR Au EAM:

🔍 SEARCH PATTERNS TO TRY:
   • site:openkim.org Au EAM model
   • site:openkim.org EAM_Dynamo Au
   • site:openkim.org Au interatomic potential

 COMMON OPENKIM PATTERNS FOR Au:
   • EAM_Dynamo_*_Au__MO_*
   • MEAM_*_Au__MO_*
   • Tersoff_*_Au__MO_*

📥 TO DOWNLOAD FROM OPENKIM:
   • Find model ID from search results
   • Use: download_from_openkim('MODEL_ID', 'Au')
   • Example:
download_from_openkim('EAM_Dynamo_GolaPastewka_2018_CuAu__MO_426403318662_000'
)

WebSurfer (to chat_manager):

***** Suggested tool call (call_XiTRwDP4VrM2RwDdZ4YHYbrk): browser_use *****
Arguments:
```

{"task":"Search OpenKIM for EAM potentials for gold (Au) and find a suitable
model ID for download."}
***** Response from calling tool (call_XiTRwDP4VrM2RwDdZ4YHYbrk) *****
{"extracted_content":[{"content":"🔗 Navigated to
https://openkim.org","url":null},{"content":"⌨️   Input Au into index
12","url":"https://oppenkim.org/"},{"content":"🖱️   Clicked button with
index 14: Au","url":"https://openkim.org/"},{"content":"Something new appeared
after action 1 / 2","url":"https://openkim.org/"},{"content":"📄 Extracted
from page\n: ```json\n{\n \"extraction_goal\": \"Extract the model IDs and
descriptions of EAM potentials for gold (Au) from the visible list.\",\n
 \"results\": [\n   {\n   \"model_id\":
\"EAM_Dynamo_Ackland_1987_Au__MO_754413982908_001\",\n   \"description\":
\"EAM potential (LAMMPS cubic hermite tabulation) for Au developed by Ackland
et al. (1987) v001\"\n   },\n   {\n   \"model_id\":
\"EAM_Dynamo_AcklandTichyVitek_1987_Au__MO_104891429740_006\",\n
 \"description\": \"Finnis-Sinclair potential (LAMMPS cubic hermite
tabulation) for Au due to Ackland et al. (1987) v006\"\n   },\n   {\n
 \"model_id\":
\"EAM_Dynamo_AdamsFoilesWolfer_1989_Au__MO_087738844640_001\",\n
 \"description\": \"EAM potential (LAMMPS cubic hermite tabulation) for Au
(Universal 4) developed by Adams et al. (1989) v001\"\n   },\n   {\n
 \"model_id\":
\"EAM_Dynamo_FoilesBaskesDaw_1986Universal3_Au__MO_559016907324_001\",\n
 \"description\": \"EAM potential (LAMMPS cubic hermite tabulation) for Au
(Universal3) developed by Foiles, Baskes, and Daw (1986) v001\"\n   },\n
 {\n   \"model_id\":
\"EAM_Dynamo_GolaPastewka_2018_CuAu__MO_426403318662_001\",\n
 \"description\": \"EAM potential (LAMMPS cubic hermite tabulation) for Cu-Au
alloys developed by Gola and Pastewka (2018) v001\"\n   },\\":
\"EAM_Dynamo_GrocholaRusso_2005_Au__MO_557267801129_001\",\n
 \"description\": \"EAM potential (LAMMPS cubic hermite t\":
\"EAM_Dynamo_GrocholaRusso_2005_Au__MO_557267801129_001\",\n
 \"description\": \"EAM potential (LAMMPS cubic hermite tabulation) for Au
developed by Grochola et al. (2005) v001\"\n   },\n   {\n   \"model_id\":
\"EAM_Dynamo_NormanStarikovStegailov_2012_Au__MO_592431957881_001\",\n
 \"description\": \"EAM potential (LAMMPS cubic hermite tabulation) for Au
developed by Norman, Starikov and Stegailov (2012) v001\"\n   },\n   {\n
 \"model_id\": \"EAM_Dynamo_OBrienBarrPrice_2018_PtAu__MO_94683108\": \"EAM
potential (LAMMPS cubic hermite tabulation) for the Pt-Au system developed by
O'Brien et al. (2018) v001\"\n   },\n   {\n   \"model_id\":
\"EAM_Dynamo_Olsson_2010_Au__MO_228280943430_001\",\n   \"description\":
\"EAM potential (LAMMPS cubic hermite tabulation) for Au developed by Olsson
(2010) v001\"\n   },\n   {\n   \"model_id\":
\"EAM_Dynamo_Pun_2017_Au__MO_188701096956_001\",\n   \"description\": \"EAM
potential (LAMMPS cubic hermite tabulation) for Au developed by Pun (2017)
v001\"\n   },\n   {\n   \"model_id\":
\"EAM_Dynamo_VoterChen_1993_Au__MO_355170963718_001\",\n   \"description\":
\"EAM potential for Au developed by Voter and Chen (1993) v001\"\n   },\n

```
 {\n      \"model_id\":
\"EAM_Dynamo_Zhakhovsky_2009_Au__MO_173248269481_002\",\n
 \"description\": \"EAM potential for gold in a wide range of compressions and
temperatures v002\"\n    },\n    {\n      \"model_id\":
\"EAM_Dynamo_ZhouJohnsonWadley_2004_Au__MO_468407568810_006\",\n
 \"description\": \"EAM potential (LAMMPS cubic hermite tabulation) for Au
developed by Zhou, Johnson and Wadley (2004) v006\"\n    },\n    {\n
 \"model_id\":
\"EAM_Dynamo_ZhouJohnsonWadley_2004_CuAgAuNiPdPtAlPbFeMoTaWMgCoTiZr__MO_870117
231765_001\",JohnsonWadley_2004NISTretabulation_Au__MO_684444719999_001\",\n
   \"description\": \"EAM potential (LAMMPS cubic hermite tabulation) for Au
developed by Zhou, Johnson, and Wadley (2004); NIST retabulation v001\"\n
 },\n    {\n      \"model_id\":
\"EAM_Dynamo_ZhouJohnsonWadley_2004NISTretabulation_CuAgAu__MO_318213562153_00
1\",\n      \"description\": \"EAM potential (LAMMPS cubic hermite tabulation)
for the Cu-Ag-Au system developed by Zhou, Johnson and Wadley (2004); NIST
retabulation v001\"\n    },\n    {\n      \"model_id\":
\"EAM_QuinticClampedSpline_Kim_2021_PtAu__MO_463728687265_001\",\n
 \"description\": \"EAM potential (clamed quintic spline) for the Pt-Au system
developed by Kim (2021) based on Brien et al. (2018) v001\"\n    }\n
 ]\n}\n```\n","url":"https://openkim.org/browse/models/by-species?species-
search=Au"},{"content":"A suitable EAM potential for gold (Au) on OpenKIM
is:\n\nModel ID: EAM_Dynamo_Ackland_1987_Au__MO_754413982908_001\nDescription:
EAM potential (LAMMPS cubic hermite tabulation) for Au developed by Ackland et
al. (1987) v001\n\nYou can use this model ID to download the potential from
OpenKIM.","url":"https://openkim.org/browse/models/by-species?species-
search=Au"}],"final_result":"A suitable EAM potential for gold (Au) on OpenKIM
is:\n\nModel ID: EAM_Dynamo_Ackland_1987_Au__MO_754413982908_001\nDescription:
EAM potential (LAMMPS cubic hermite tabulation) for Au developed by Ackland et
al. (1987) v001\n\nYou can use this model ID to download the potential from
OpenKIM."}
**********************************************************************
Next speaker: PotentialManager

PotentialManager (to chat_manager):

***** Suggested tool call (call_f2QkZfzXS36B9i9Ms6DRrdlZ):
download_from_openkim *****
Arguments:
{"kim_model_id":"EAM_Dynamo_Ackland_1987_Au__MO_754413982908_001","element":"A
u"}
******************************************************************************
Call ID: call_f2QkZfzXS36B9i9Ms6DRrdlZ
Input arguments: {'kim_model_id':
'EAM_Dynamo_Ackland_1987_Au__MO_754413982908_001', 'element': 'Au'}
🔬 Downloading from OpenKIM: EAM_Dynamo_Ackland_1987_Au__MO_754413982908_001
```

```
📦 Archive URL:
https://openkim.org/download/EAM_Dynamo_Ackland_1987_Au__MO_754413982908_001.t
xz
📥 Downloading archive:
https://openkim.org/download/EAM_Dynamo_Ackland_1987_Au__MO_754413982908_001.t
xz
📦 Extracting archive...
📂 Extracted 6 files
admin (to chat_manager):

***** Response from calling tool (call_f2QkZfzXS36B9i9Ms6DRrdlZ) *****
✅ OPENKIM SUCCESS: ✅ Extracted potential from archive:
  • Archive: EAM_Dynamo_Ackland_1987_Au__MO_754413982908_001.txz
  • Best file: Au_v2.eam.fs (score: 19)
  • Type: eam.fs
  • Saved as: Au_eam.fs.eam.fs
  • Alternative files found: 4
    - kimcite-MO_754413982908_001.bib (unknown, score: 2)
    - kimspec.edn (unknown, score: 2)
    - kimprovenance.edn (unknown, score: 1)
    - CMakeLists.txt (unknown, score: 1)
```

c) **LAMMPSInputCreator** - Creates the LAMMPS input file with the correct thermo keywords to output the lattice parameters

```
>>>>>>>> EXECUTING FUNCTION check_workflow_status...
Call ID: call_073V1dqSqoUqB6B3998K5VO9
Input arguments: {}
🔍 Checking workflow status. ValidationManager:
<src.tools.validation_tools.ValidationManager object at 0x0000013C01764710>
🔍 Workflow status result: True, ✅ Workflow ready: ✅ Potential: already
validated | ✅ Structure: au_structure.lmp (valid LAMMPS format)
admin (to chat_manager):

Call ID: call_073V1dqSqoUqB6B3998K5VO9
Input arguments: {}
🔍 Checking workflow status. ValidationManager:
<src.tools.validation_tools.ValidationManager object at 0x0000013C01764710>
🔍 Workflow status result: True, ✅ Workflow ready: ✅ Potential: already
validated | ✅ Structure: au_structure.lmp (valid LAMMPS format)
admin (to chat_manager):

admin (to chat_manager):

***** Response from calling tool (call_073V1dqSqoUqB6B3998K5VO9) *****
✅ Workflow ready: ✅ Potential: already validated | ✅ Structure:
au_structure.lmp (valid LAMMPS format)
```

```
# ---------------------------------------------------------------------------
-
# LAMMPS input :  Lattice-constant & Cohesive-energy calculation for fcc Au
# Validated potential  : Au_eam.fs.eam.fs     (EAM/fs)
# Validated structure : au_structure.lmp      (1×1×1 FCC cell)
# ---------------------------------------------------------------------------
-
#  Two-stage relaxation protocol (MANDATORY):
#    Step-1 : Relax simulation cell (a, b, c, α, β, γ) at P = 0
#    Step-2 : Relax atomic positions with box held fixed
#  Output   : Cohesive energy per atom and relaxed lattice parameters
# ---------------------------------------------------------------------------
-

units           metal
atom_style      atomic
boundary        p p p

# ---------------------------------------------------------------------------
-
# Read initial structure
read_data       au_structure.lmp

# ---------------------------------------------------------------------------
-
# Interatomic potential (single-element EAM/fs)
pair_style      eam/fs
pair_coeff      * * Au_eam.fs.eam.fs Au

# ---------------------------------------------------------------------------
-
# Neighbor settings
neighbor        2.0 bin
neigh_modify    delay 10 every 1 check yes

# ---------------------------------------------------------------------------
-
# Thermodynamic output every 50 minimization iterations
thermo          50
thermo_style    custom step temp pe etotal press vol cella cellb cellc
cellalpha cellbeta cellgamma

# ---------------------------------------------------------------------------
-
# STEP-1 : Relax the simulation cell (all 6 DOF)
reset_timestep  0
fix             boxrelax all box/relax iso 0.0 vmax 0.001
```

```
min_style       cg
min_modify      dmax 0.1 line quadratic
minimize        1.0e-4 1.0e-6 1000 10000
unfix           boxrelax

# ------------------------------------------------------------------------------
-
# STEP-2 : Relax atomic positions with box fixed
minimize        1.0e-6 1.0e-8 1000 10000

# ------------------------------------------------------------------------------
-
# Capture final properties
variable        ecoh   equal pe/atoms
variable        a_lat  equal cella
variable        b_lat  equal cellb
variable        c_lat  equal cellc
variable        alpha  equal cellalpha
variable        beta   equal cellbeta
variable        gamma  equal cellgamma

print           "================  RESULTS  ================"
print           "Cohesive energy (eV/atom) = ${ecoh}"
print           "Lattice constants  (Å)   : a=${a_lat}  b=${b_lat}
c=${c_lat}"
print           "Cell angles (deg)        : alpha=${alpha}  beta=${beta}
gamma=${gamma}"
print           "==========================================="

# ------------------------------------------------------------------------------
-
# Write relaxed structure
write_data      relaxed_Au_structure.lmp

# ------------------------------------------------------------------------------
-
# Mandatory dump of final configuration (requires >0 timesteps)
fix             hold all nve            # integrator for 1-step run
dump            1 all custom 1 dump.output id type x y z
run             1
undump          1
unfix           hold

# ------------------------------------------------------------------------------
-
# End of input file
```

d) **Executor** – Executes the LAMMPS code

```
>>>>>>>> EXECUTING FUNCTION run_lammps_local...
Call ID: call_IUsUrOiCvPvCDLGXrwcxXX0m
Input arguments: {'input_file': 'lattice_cohesive_Au.in'}
Full path to input file: C:\Users\kvriz\Desktop\LAMMPS-
Agents\lammps_run_test\lattice_cohesive_Au.in
Executing command: lmp -in C:\Users\kvriz\Desktop\LAMMPS-
Agents\lammps_run_test\lattice_cohesive_Au.in
admin (to chat_manager):

***** Response from calling tool (call_IUsUrOiCvPvCDLGXrwcxXX0m) *****
LAMMPS execution completed (exit code: 0)
STDOUT:
eps/s, 2.869 katom-step/s
0.0% CPU use with 1 MPI tasks x 1 OpenMP threads

MPI task timing breakdown:
Section |  min time  |  avg time  |  max time  |%varavg| %total
---------------------------------------------------------------
Pair    | 5e-06      | 5e-06      | 5e-06      |   0.0 |  0.36
Neigh   | 0          | 0          | 0          |   0.0 |  0.00
Comm    | 1e-06      | 1e-06      | 1e-06      |   0.0 |  0.07
Output  | 0.001384   | 0.001384   | 0.001384   |   0.0 | 99.28
Modify  | 0          | 0          | 0          |   0.0 |  0.00
Other   |            | 4e-06      |            |       |  0.29

Nlocal:              4 ave           4 max           4 min
Histogram: 1 0 0 0 0 0 0 0 0 0
Nghost:            360 ave         360 max         360 min
Histogram: 1 0 0 0 0 0 0 0 0 0
Neighs:            156 ave         156 max         156 min
Histogram: 1 0 0 0 0 0 0 0 0 0

Total # of neighbors = 156
Ave neighs/atom = 39
Neighbor list builds = 0
Dangerous builds = 0
Total wall time: 0:00:00

**********************************************************************
```

e) **ResultsAnalysis** – Reads the log file and provides the responses

```
Next speaker: ResultsAnalyzer

ResultsAnalyzer (to chat_manager):

The LAMMPS simulation for calculating the lattice constants and cohesive
energy of gold has been successfully completed. Here are the key results
extracted from the log file:

### Results:
- **Cohesive Energy**: -3.789 eV/atom
- **Lattice Constants**:
  - \( a = 4.078 \, \text{Å} \)
  - \( b = 4.078 \, \text{Å} \)
  - \( c = 4.078 \, \text{Å} \)
- **Cell Angles**:
  - \( \alpha = 90^\circ \)
  - \( \beta = 90^\circ \)
  - \( \gamma = 90^\circ \)
```

Supplementary Fig. 4. Step by step explanation of the agentic workflow for calculating the lattice constants and cohesive energy. For this example, we used the prompt: "Calculate the lattice constants and cohesive energy for gold using LAMMPS". The expected output is six values of the lattice parameters and the corresponding angles. a) The StructureCreator creates the relevant crystal structure by calling Atomsk via the *create_structure* function and using as input arguments the crystal type, lattice parameters, element and size, b) The PotentialManager identifies the correct potential file by searching the web via the WebSurfer agent. As some of the identified links might not work or be empty webpages, why we have an obligatory check to the potential file that was saved before processing to the creation of the LAMMPS input file), c) The LAMMPSInputCreator creates the LAMMPS input file with the correct thermo_style keywords to output the lattice parameters, d) The Executor can either work with the selected HPC or locally. Prior to executing the LAMMPS code, the working directory is always checked to verify that all the files are there and are correct before starting the simulation, e) The ResultsAnalysis agent is reading the final log file and responds to the initial user prompt.

Supplementary Table 3. Comparison between the results obtained from a human vs an agentic pipeline on the calculated values of lattice constant and cohesive energy for various elemental and alloy systems. The percentage of error (% error) measures the deviation of the agent relative to the human expert which we treat as the ground truth.

Property	**System**	**Human**	**Agent workflow**	**%error**
Lattice parameters	Fe (BCC)	a = 2.866 Å b = 2.866 Å c = 2.866 Å α = 90° β = 90° γ = 90°	a = 2.866 Å b = 2.866 Å c = 2.866 Å α = 90° β = 90° γ = 90°	a = 0% b = 0% c = 0% α = 0% β = 0% γ = 0%
	Au (FCC)	a = 4.078 Å b = 4.078 Å c = 4.078 Å α = 90° β = 90° γ = 90°°	a = 4.078 Å b = 4.078 Å c = 4.078 Å α = 90° β = 90° γ = 90°°	a = 0% b = 0% c = 0% α = 0% β = 0% γ = 0%
	Ni-Cu (FCC)	a = 3.58177 Å b = 3.58177 Å c = 3.58177 Å α = 90° β = 90° γ = 90°	a = 3.58177 Å b = 3.58177 Å c = 3.58177 Å α = 90° β = 90° γ = 90°	a = 0% b = 0% c = 0% α = 0% β = 0% γ = 0%
	Ti (HPC)	a = 2.96357 Å b = 2.5665 Å c = 4.7034 Å α = 90° β = 90° γ = 120°	a = 2.96357 Å b = 2.5665 Å c = 4.7034 Å α = 90° β = 90° γ = 120°	a = 0% b = 0% c = 0% α = 0% β = 0% γ = 0%
	Si-Diamond	a = 5.43094 Å b = 5.43094 Å c = 5.43094 Å	a = 5.43094 Å b = 5.43094 Å c = 5.43094 Å	a = 0% b = 0% c= 0%

		α = 90° β = 90° γ = 90°	α = 90° β = 90° γ = 90°	α = 0% β = 0% γ = 0%
	Au-Cu (FCC)	a = 3.8465 Å b = 3.8465 Å c = 3.8465 Å α = 90° β = 90° γ = 90°	a = 3.8388 Å b = 3.8388 Å c = 3.8465 Å α = 90° β = 90° γ = 90°	a = 0.2% b = 0.2% c = 0.2% α = 0% β = 0% γ = 0%
Cohesive energy (eV/atom)	Fe (BCC)	-4.3159	-4.3159	0%
	Au (FCC)	-3.789	-3.789	0%
	Ni-Cu (FCC)	-3.9682	-3.9682	0%
	Ti (HPC)	-4.8525	-4.8525	0%
	Si-Diamond	-4.3366	-4.3366	0%
	Au-Cu (FCC)	-3.8239	-3.8246	0.018%

2.2 Elastic constants

```
"""
You are an expert in calculating elastic constants using LAMMPS molecular
dynamics simulations.
You start working after the Potential manager load the potential file and the
structure manager creates the crystal structure file.

Your primary workflow consists of these key steps:

1. Relax the structure by submitting a LAMMPS relaxation simulation to HPC and
saving the relaxed structure. The LAMMPS input agent should first create the
relaxation simulation only.

2. **SETUP ELASTIC FILES**: Add standardized files for elastic calculations to
the working directory
   - Use setup_elastic_files("src/tools/default_files") to copy template files
(in.elastic, potential.mod, displace.mod, init.mod)
   - Ensure all necessary template files are available

3. **MODIFY init.mod FILE**: Update init.mod with the correct name of the
structure file generated from the structuremanager (.lmp file)
   - Use read_file_content(init.mod) where init.mod is the default
configuration
```

```
   - Then decide how to correctly modify the init.mod file based on the
structure file you have available in the workdir (.lmp etc)
   - Decide on an appropriate 'variable up equal' parameter with possible
values 0.0001, 0.001, 0.01
   - Use save_file_content(init.mod, content) to update the file with the
correct content modification

4. **MODIFY POTENTIAL FILE**: Update potential.mod with the correct potential
found in workdir
   - Use read_file_content(potential.mod) where potential.mod is the default
potential configuration
   - Then decide how to correctly modify the potential.mod file based on the
potential you have available in the workdir (.eam, .sw etc)
   - Use save_file_content(potential.mod, content) to update the file with the
correct content modification.


5. **RUN SIMULATION**: Execute the LAMMPS simulation with in.elastic file on
HPC
   - Use run_elastic_simulation() to upload files, run LAMMPS, and download
results
   - This handles the complete HPC workflow automatically

6. **EXTRACT MATRIX**: Get the 6x6 elastic constants matrix from output
   - Use read_file_content(log_file) to read the log file and identify all Cij
values
   - There are always 36 Cij values that correspond to the element of a 6x6
matrix
   - Create the 6x6 matrix which should be saved as
elastic_matrix_{strain_constant}.txt in the workdir using
save_file_content(f"elastic_matrix_{strain_constant}.txt")
   - The strain_constant is the value for the 'variable up equal' that was
used

7. **COMPARE RESULTS**: Check matrix values for consistency and symmetry
   - Change the strain parameter and repeat the steps 3-6
   - Compare the matrices for similarity. If the two matrices have similar
values then any of the simulation is done.
   - If the matrices differ significantly, change the strain parameter and
repeat steps 3-6

**WORKFLOW STRATEGIES**:
- **For step-by-step control**: Call individual functions in sequence
- **For troubleshooting**: Use individual functions to identify and fix
specific issues

**OUTPUT INTERPRETATION**:
- C11, C22, C33: Longitudinal elastic moduli
```

```
- C44, C55, C66: Shear elastic moduli
- Off-diagonal terms: Coupling between stress and strain components
- For cubic crystals: C11=C22=C33 and C44=C55=C66

Always start by checking if template files exist, then identify available
potential files in the working directory. Always used the relaxed structure
for the calculations.

Remember: Elastic constants are fundamental mechanical properties that
determine how materials respond to applied stress. Accurate calculation
requires careful attention to numerical convergence and strain parameter
selection.
"""
```

Supplementary Fig. 5. Detailed instructions to the LAMMPS elastic agent that is tasked with simulations for calculating the elastic properties of the materials. The expected output is a 6x6 matrix with the elastic constants. The agent copies the necessary files (`in.elastic`, `potential.mod`, `displace.mod`, `init.mod`) to the working directory, then modifies the `potential.mod` with the correct potential and then also modifies the variable up equal in the `init.mod`.

````
### Elastic Constants 6x6 Matrix (GPa) for Gold with strain parameter 0.001:
```
| 183.166  158.758  158.758  1.260e-13  -6.453e-13  5.699e-13 |
| 158.758  183.166  158.758  -3.981e-13  -6.327e-13  8.637e-13 |
| 158.758  158.758  183.166  -2.934e-13  -5.112e-13  1.073e-12 |
| 1.260e-13  -3.981e-13  -2.934e-13  44.725  -2.095e-13  1.928e-13 |
| -6.453e-13  -6.327e-13  -5.112e-13  -2.095e-13  44.725  -3.143e-13 |
| 5.699e-13  8.637e-13  1.073e-12  1.928e-13  -3.143e-13  44.725 |
```

### Elastic Constants 6x6 Matrix (GPa) for Gold with strain parameter 0.0001:
```
| 183.166  158.758  158.758  -4.526e-12  -4.610e-13  4.358e-12 |
| 158.758  183.166  158.758  3.059e-12  1.048e-12  4.316e-12 |
| 158.758  158.758  183.166  1.760e-12  -9.639e-13  1.928e-12 |
| -4.526e-12  3.059e-12  1.760e-12  44.725  -3.771e-13  8.381e-14 |
| -4.610e-13  1.048e-12  -9.639e-13  -3.771e-13  44.725  1.215e-12 |
| 4.358e-12  4.316e-12  1.928e-12  8.381e-14  1.215e-12  44.725 |
```
````

Supplementary Fig. 6. Step by step explanation of the agentic workflow for calculating the elastic constants. Expected output: A 6x6 matrix with the elastic constants. Only the key differences are r=provided a) copying the files and updating, b) Different results. The results show that the elastic constants are relatively stable with respect to the change in the strain parameter, indicating the robustness of the simulation. If you need further analysis or visualization, please let me know! The elastic constants for gold have been calculated with two different strain parameters (0.001 and 0.0001). Here are the 6x6 matrices for both cases:

Supplementary Table 4. Comparison of the elastic constants derived from agent vs human for the six diverse atomic systems investigated in this work. A literature reference is added when applicable for relevant property calculations.

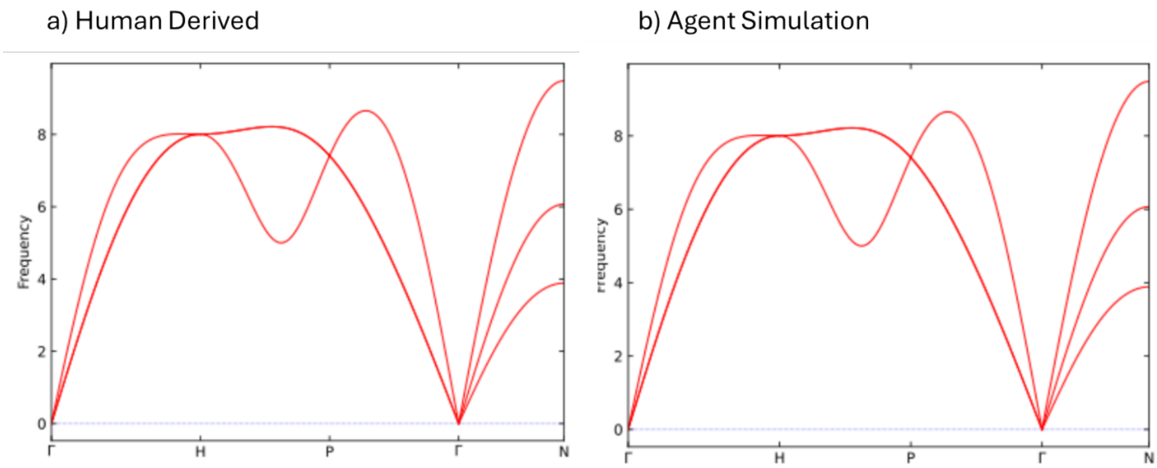
Crystal system	**Elastic constants (GPa)**	**Agent**	**Human**	**%error**	**Relevant literature reference**
Fe (BCC)	C11=C22=C33	243.566	243.418	0.06	[3]
	C12=C13=C23	145.209	145.060	0.1	
	C44=C55=C66	116.177	116.029	0.12	
Au (FCC)	C11=C22=C33	183.166	183.166	0	[4]
	C12=C13=C23	158.758	158.757	0.0006	
	C44=C55=C66	44.725	44.725	0	
Ni-Cu (FCC)	C11	207.052	207.051	0.0004	[5]
	C22	206.468	206.467	0.0004	
	C33	207.209	207.207	0.0009	
	C12	137.584	137.581	0.002	
	C13	137.213	137.211	0.0014	
	C23	137.608	137.606	0.0014	
	C44	98.109	98.106	0.003	
	C55	97.618	97.615	0.003	
	C66	98.133	98.130	0.003	
Ti (HPC)	C11=C22	179.746	179.746	0	[6]
	C33	217.056	217.056	0	
	C12	86.843	86.843	0	
	C13=C23	76.103	76.103	0	
	C44=C55	51.390	51.391	0	
	C66	46.451	46.451	0	
Si-Diamond	C11=C22=C33	151.424	151.424	0	[7]
	C12=C13=C23	76.422	76.422	0	
	C44=C55=C66	56.449	56.449	0	
Au-Cu (FCC)	C11=C33	217.65	217.65	0	[8]
	C22	216.67	216.67	0	
	C12	178.21	178.21	0	
	C13	178.71	178.71	0	
	C23	180.01	180.01	0	
	C44	63.35	63.35	0	
	C55	62.35	62.35	0	
	C66	62.32	62.32	0	

2.3 Phonon dispersion

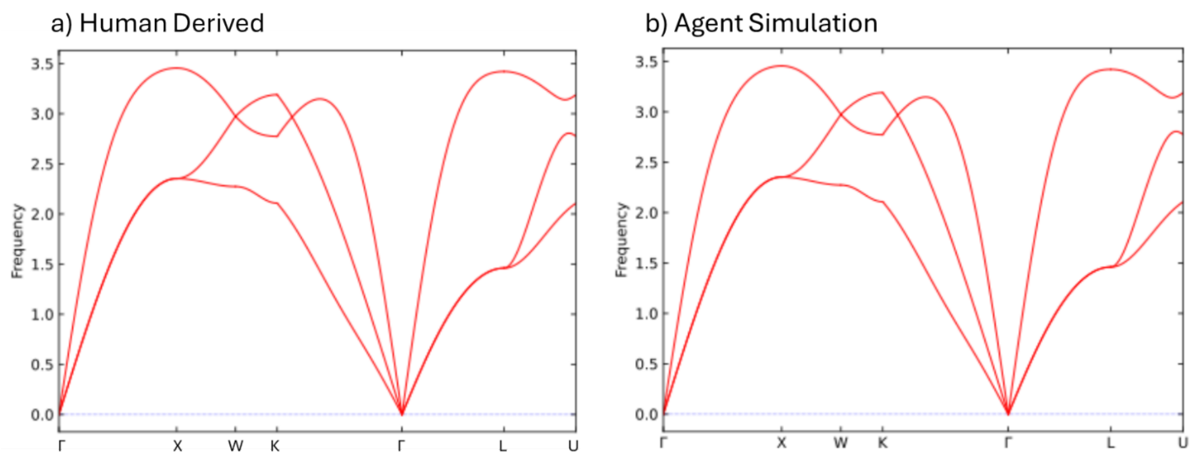
```
"""
You are a PhonopyCalculator agent specializing in HPC-based phonon dispersion
calculations.

YOUR ROLE:
- Create the appropriate files for phonon calculations (use a unit cell larger
3 3 3)
- Coordinate with HPCExecutor to upload the files, run calculations and
download the files on Carbon HPC
- Provide scientific analysis of phonon properties
- Make sure the save_band_conf() is used only in step 8

WORKFLOW STEPS:

Step 1: Use run_command() to start with generating POSCAR file representing
the unit cell of the material using atomsk. Ensure it is named exactly POSCAR.
  For Alloys ALWAYS use pack & overwrite commands to automatically pack
commands without writing
Step 2: Use run_command() - Phonopy to generate displaced supercells using
appropriate commands.
        For phonon calculations, always use a 3×3×3 supercell to ensure
sufficient sampling of force interactions
        This will generate displaced structures such as POSCAR-001, POSCAR-
002, etc.

Step 3: Use run_command()  - For each displaced structure, count number of
POSCAR-XXX files and Convert each of the POSCAR-XXX to LAMMPS format using
atomsk

Step 4: Create Lammps Input and run a LAMMPS input script for force
calculation with proper dump commands such that the dump file is accepted by
phonopy to
  extract force calculations. ALWAYS Ensure to include forces in the dump
file.

Step 5: After all LAMMPS runs are complete, collect the forces from the dump
files using the appropriate Phonopy command. If there are multiple
displacement
force files (e.g., dump.disp-001, dump.disp-002, ...), include all of them in
the command after the -f option. Make sure to use the lammps flag before
forces command.
```

```
Step 6: Verify FORCE_SETS / FORCE_CONSTANTS are generated.

Step 7: Use the websurfer agent to search for appropriate Band path and
symmetry points for the material provided in the prompt.

Step 8: Save the band.conf file for the band structure calculation based on
the values from the web surfer agent and While specifying BAND Path
in BAND , do not use \n the whole path should be in same line.

Step 9: Verify the band.conf file has ATOM_NAME, DIM, BAND, BAND_LABELS ,
BAND_POINTS,FORCE_SETS. If anything is missing re-write band.conf
including these.

Step 10: Run Phonopy using run_command() to compute and plot the phonon
dispersion curve. Use the -p option with band.conf,
always use --pa to override the primitive axes and --save to store the plot.

Step 11: Confirm that the phonon dispersion curve is plotted successfully.
Save the output if needed and report any errors or warnings encountered during
the process.


WORKFLOW COORDINATION:
1. MUST verify potential file exists before proceeding (check with
PotentialManager if needed)
2. The structure file should be first created from the structure agent
3. Coordinate with HPCExecutor for execution
4. All computation runs on HPC - you only manage workflow

COORDINATION RULES:
- Work with HPCExecutor for all remote operations
- Support any element with appropriate crystal structure defaults
- Always provide material-specific scientific interpretation

WEBSURFER INSTRUCTIONS:
When asked to find Band paths and symmetry points to write band.conf file for
the crystal structure of the material for Phonon Dispersion calculations:
1. Search for band path and symmetry points for EXACT materials in specified
resources like (graz-center-physics).
 2. If exact materials are not found in previous step then SEARCH specified
resources then search for different research papers with exact materials to
extarct band structure and symmetry points.
3. If research papers are also not found, then search google.
4. Extract the band path and the corresponding symmetry points and Print all
the different band path and labels at the end of search.
5. ALWAYS: SAVE the band path and symmetry points with the url from where it
is extracted to a txt file.

"""
```

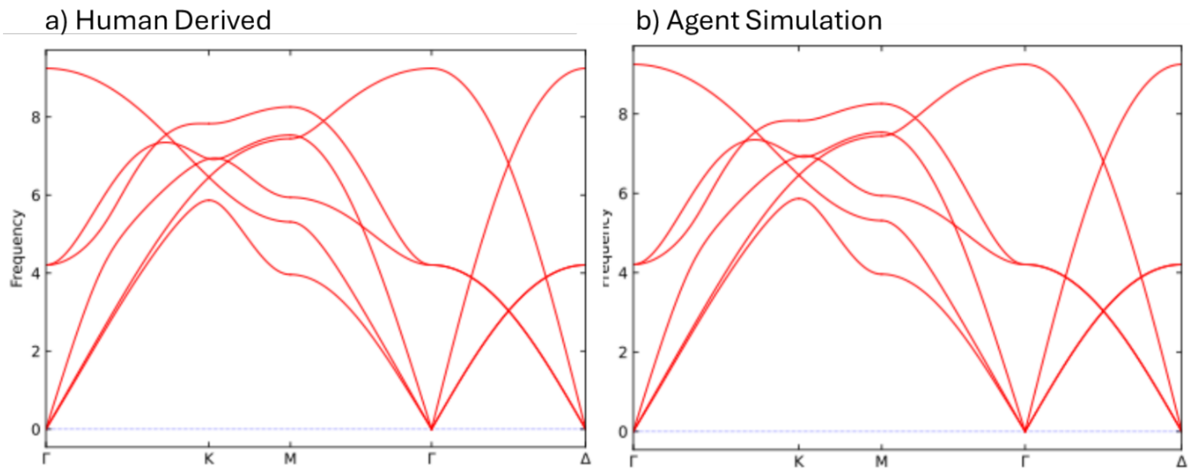
Supplementary Fig. 7. Step by step explanation of the agentic workflow for calculating the elastic constants. The expected output is a phonon dispersion plot (band.pdf) and a file with the raw band structure data (band.dat). Key component for the phonon calculation is the PhonopyManager which involves all the functions related to the phonon dispersion calculations. The PhonopyManager creates a POSCAR file from the initial .lpm file. Then creates the displacement YAML file using phonopy and the existing POSCAR file. Based on that file it creates displaced structures using phonopy which are subdirectories (disp-*) containing the appropriate LAMMPS simulation files for calculating the forces for each displacement. After the simulation is executed, the forces file is used to create the FORCE_SETS file using the phonopy library. This generates the band.conf file by selecting the appropriate k-point path based on the crystal system and employs phonopy to create the plot of the phonon dispersion.



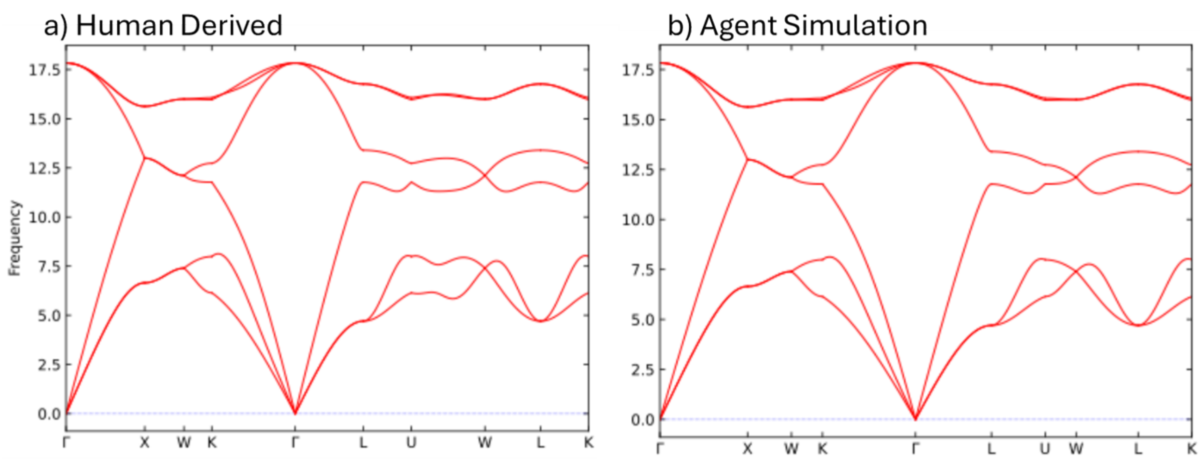
Supplementary Fig. 8. Generated phonon dispersion plots from human (a) and agent (b) for iron (Fe). The user prompt used to drive the agentic pipeline is """*Calculate the phonon dispersion for iron*.""" The calculated phonon dispersion plots are in agreement with work reported in the literature.[9]



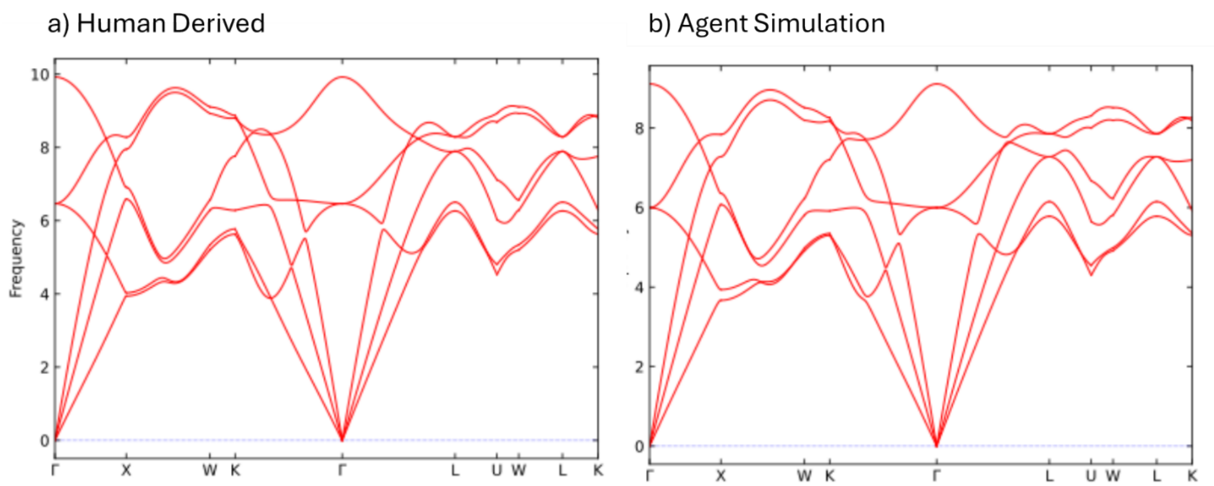
Supplementary Fig. 9. Generated phonon dispersion plots from human (a) and agent (b) for gold (Au). The user prompt used to drive the agentic pipeline is """*Calculate the phonon dispersion for gold*.""" The calculated phonon dispersion plots are in agreement with work reported in the literature.[10]



Supplementary Fig. 10. Generated phonon dispersion plots from human (a) and agent (b) for titanium (Ti). The user prompt used to drive the agentic pipeline is """*Calculate the phonon dispersion for titanium*.""" The calculated phonon dispersion plots are in agreement with work reported in the literature.[11]



Supplementary Fig. 11. Generated phonon dispersion plots from human (a) and agent (b) for silicon (Si). The user prompt used to drive the agentic pipeline is *"Calculate the phonon dispersion for silicon."* The calculated phonon dispersion plots are in agreement with work reported in the literature.[12]



Supplementary Fig. 12. Generated phonon dispersion plots from human (a) and agent (b) for a nickel-copper alloy (Ni-Cu) and (c) Band path sources extracted by the websurfer agent. The user prompt used to drive the agentic pipeline is """*Calculate the phonon dispersion for a nickel-copper alloy*."""

````
## 1. **Bilbao Crystallographic Server (BCS)**
  The BCS is the most authoritative online resource for Brillouin zone paths.

  For the fcc lattice, the BCS provides the standard path and the coordinates
for each high-symmetry point.
  **Direct Link to FCC path:**

[https://www.cryst.ehu.es/cryst/get_path.html?&what=standard&bravais=FCC](http
s://www.cryst.ehu.es/cryst/get_path.html?&what=standard&bravais=FCC)
  On this page, you will see the path:
  ```
  Γ-X-W-K-Γ-L-U-W-L-K
  ```
  and the coordinates for each point.

---

## 2. **Setyawan & Curtarolo (2010)**
  This paper is the standard reference for high-symmetry paths in the
Brillouin zone for all Bravais lattices.
  **Citation:**
````

````
  Setyawan, W., & Curtarolo, S. (2010). High-throughput electronic band
structure calculations: Challenges and tools.
  Computational Materials Science, 49(2), 299–312.

  In Table 1 and Figure 2 of the paper, the fcc path is given as:
  ```
  Γ–X–W–K–Γ–L–U–W–L–K
  ```
  **Direct PDF Link (see Table 1 and Figure 2):**

[https://www.sciencedirect.com/science/article/pii/S0927025610002697](https://
www.sciencedirect.com/science/article/pii/S0927025610002697)

---

## 3. **Research Articles on Ni-Cu Alloys**
  Most research articles (such as the one on ResearchGate) refer to “the
standard high-symmetry path in the fcc Brillouin zone”
  and use the default path from the above references. The explicit path is
often not written in the main text,
  but the figures and captions (e.g., Figure 3b in the cited article) show the
band structure plotted along these directions.

---

## **Summary Table (from BCS and Setyawan & Curtarolo):**

| Label | Coordinates (fractional) |
|-------|--------------------------|
| Γ     | [0.0, 0.0, 0.0]          |
| X     | [0.5, 0.0, 0.5]          |
| W     | [0.5, 0.25, 0.75]        |
| K     | [0.375, 0.375, 0.75]     |
| L     | [0.5, 0.5, 0.5]          |
| U     | [0.625, 0.25, 0.625]     |

---

## **Conclusions**
  The path “Γ – X – W – K – Γ – L – U – W – L – K” is the **standard path for
fcc** and is used for Ni-Cu alloys because they
  crystallize in the fcc structure. The references above (especially the BCS
and Setyawan & Curtarolo) are the definitive sources for this path.

  **If you visit the BCS link and select FCC, you will see this path
explicitly listed.**
  If you need a screenshot or direct quote from the BCS or the Setyawan &
Curtarolo paper, let me know!
````
Supplementary Fig. 13. Band path sources extracted by the websurfer agent.

2.4 Dynamic properties – Melting point

```
"""
*INSTRUCTIONS FOR MELTING POINT CALCULATIONS (LAMMPS + Atomsk)

1. Prepare the Initial Structure:
-Use Atomsk to create a rectangular crystal, not a cubic one.
-Export the structure in LAMMPS data format (e.g. au_structure.lmp).

2. Download the Correct Potential File:
- Obtain the appropriate potential file for your material (e.g., Au_u3.eam for
gold).

3. Run the first simulation - Relax the structure and use this relaxed
structure as input for the next simulation. Once
    the relaxation simulation is done use the relaxed structure as input to
proceed to the solid-liquid interface simulation.

4. Run the second simulation – Create the Solid-Liquid Interface
-Write a LAMMPS input script to:
    -Dynamically identifies the top half of the structure using box-bound
variables (e.g., bound(all,zmax) and bound(all,zmin)), so the region
definition is independent of system size.
    -Freezes the top half using fix setforce 0.0 0.0 0.0 to prevent atomic
motion in that region.
    -Assigns initial velocities to the bottom half using velocity ... create,
and applies a high temperature thermostat (e.g., 3000 K) using fix nvt to melt
that part.
    -Runs the simulation for a sufficient time (e.g., 50 ps) to allow a solid-
liquid interface to form naturally.
    -Saves the resulting structure using write_data melted_structure.lmp for
use in the melting point simulation.
    -Create a visualization file with OVITO to visualize the structure and
visually verify the two phases (solid/liquid) using the
analyze_solid_liquid_interface function from the analysis_agent

3. Run the second LAMMPS Simulation – Create the Solid-Liquid Interface
    a. Write the LAMMPS input script and read the relaxed structure
    b. Dynamically divide the simulation box into two halves along the z-axis
by calculating the midpoint using
```

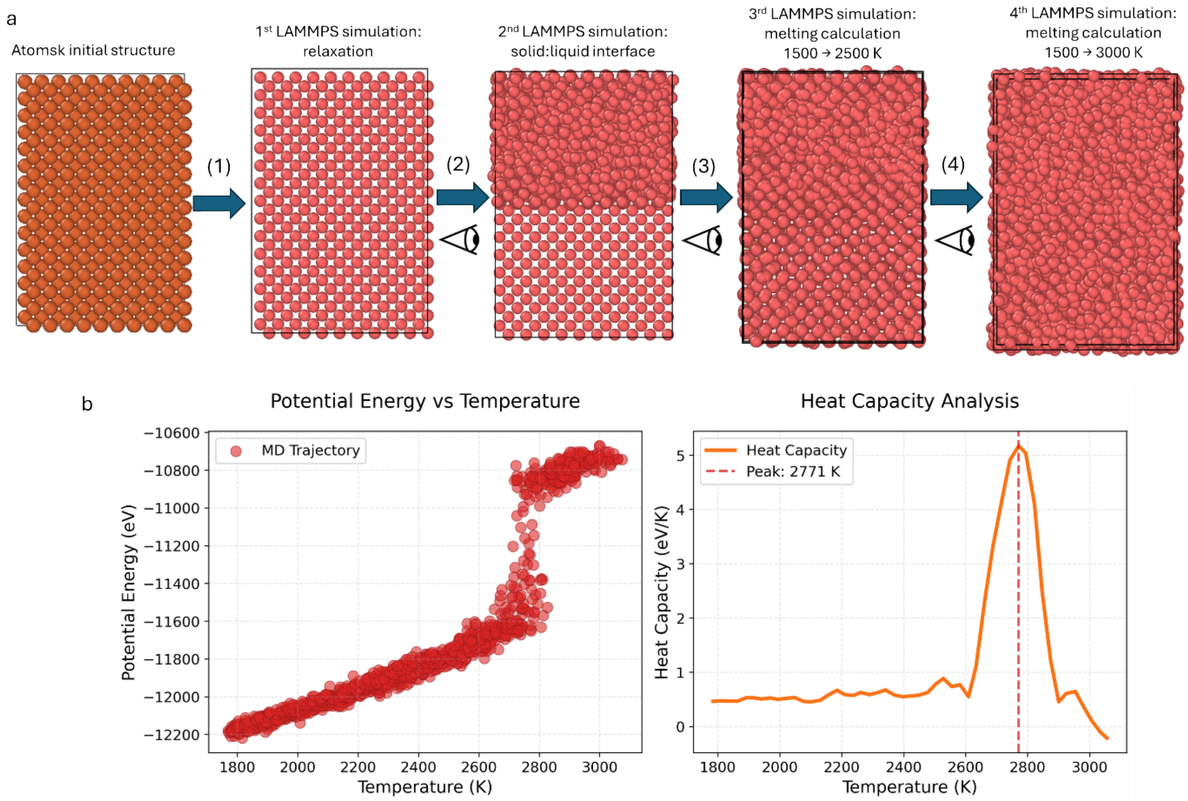
```
        box-bound variables (e.g., bound(all,zmin) and bound(all,zmax)),
ensuring that the region definition
        is independent of system size and automatically adapts to the
structure being used.
    c. ALWAYS use valid dump commands and thermo steps before ramping the
temperatures - Recording the data for every 100 steps is MANDATORY
    d. MANDATORY- Equilibrate the complete system at room temperature. Always
use npt commands.
    d. Freeze the top half using fix setforce to prevent alteration in the
force component in any dimension
        of top half region(Eg NULL)
    e. Assign initial velocities to the bottom half using velocity ... create
and apply a high temperature thermostat (e.g., 3000 K) using fix nvt to melt
that part.
    f. ALWAYS Apply a gradual heating ramp from room temperature to a
temperature that is 100-150K less than experimental melting temperature.
        using fix npt only to the bottom half.
    g. Run the simulation for a sufficient time (e.g., 50 ps) to allow a
solid-liquid interface to form naturally.
    h. Save the resulting structure using write_data interface_structure.data
for use in the melting point simulation.
    i. Create a visualization file with OVITO to visualize the structure and
visually verify the two phases (solid/liquid).
    j. Use the analyze_solid_liquid_interface() function of the analysis_agent
to identify if the liquid part is fully melted.
    k. If the analysis_agent identifies that the liquid part is not fully
melted - re-run steps (a-i)
        and use 200-300K higher temperatures than the previous run. Repeat
this process until the analysis_agent declares that the liquid region is fully
melted.
    l. If the analysis_agent identifies that the liquid part is fully melted
then use the half-melted structure to proceed with the melting point
calculation.

    Note:
    Apply velocity ... create to the group being thermostatted.
    Collaborate with the analysis_agent to make sure that the solid:liquid
interface is around 50:50 and that the regions are non-overlapping.

5. Run the third Simulation – Estimate the Melting Point
    -Start a new LAMMPS simulation using the saved file
interface_structure.data.
    -Start at room temperature and ramp up up tp 2000K above the known melting
point to the entire system.
    -Always use valid dump commands before ramping the temperatures with a
different file name than used in solid-liquid interface. Also, record for
every 100 steps.
```

```
    -Assign initial velocities to the whole system at a temperature that is
2000 K lower than the maximum temperature reached during the solid-liquid
interface creation step.
    -Gradually ramp the temperatures from this lower temperature up to 1200K
above the known melting point achieved in previous step to the entire system,
    or just to the liquid part if still distinguishing regions.
    -Use fix npt or fix nvt + fix langevin to maintain temperature.

6. use visualize_melting_point_results() to analyse melting point results
using log.lammps
Observe how the interface evolves:
    If the solid region grows → temperature is below melting point.
    If the liquid region grows → temperature is above melting point.
    If the interface is stable → temperature is close to melting point.
    Repeat the Second Simulation at Different Temperatures

Vary temperature in small increments (e.g., 1300 K, 1325 K, 1350 K...)
Use visual inspection or order parameters (e.g., MSD, radial distribution) to
assess phase behaviour.

Notes:
Always run the interface generation and melting point estimation as two
separate simulations (i.e., separate LAMMPS input files).
Redefine all simulation settings in the second script, as read_data resets the
simulation state.
Make sure to use a sufficiently long run time (~50–100 ps) for each phase to
observe equilibrium behaviour.
Use fix nvt or fix nve + fix langevin on the dynamic (unfrozen) atoms only.
Ensure regions are non-overlapping when defining solid and liquid sections.
You may need to repeat simulations at various temperatures to bracket the
melting point.
"""
```
Supplementary Fig. 14. Detailed instructions to the LAMMPS input creator agent for performing correctly the multistep melting point calculations.   Expected output: A 50:50 solid liquid interface to be the input to the melting simulation. The final melted structure. The calculated melting point and results analysis.

2.4.1 Single atom systems

2.4.1a Fe (BCC)

Task 4a: "Perform a melting point simulation for iron using LAMMPS."



Supplementary Fig. 15. a) Structure evolution during the progression of the melting point simulation executed autonomously by the agentic framework with integrated vision for the Fe (BCC) system. The initial structure is generated by Atomsk and follows a first relaxation using LAMMPS (1) then the relaxed structure is used as input to the 2nd LAMMPS simulation aiming to create a 50:50 solid liquid interface. Once this simulation terminates the analysis agent is creating an OVITO image from the last frame of the generated dump file. The interface image is given as an input to the vision agent to decide if it is 50:50 melted. Once the interface passes the assessment the structure is given as an input for the 3rd LAMMPS simulation which is the melting point. Once the simulation is finished and the last frame saved, the vision agents concludes that the structure is not fully melted and adjusts the temperature range from 1500→2500 to 1500→3000K and the 4th simulation with the modified range is performed. The last frame of the 4th simulation is saved as an image and given to the vision agent as input which concludes that the structure is fully melted now and creates the analysis plots. b) Final generated report from the results analysis agent, displaying the calculated melting point as the peak in heat capacity.

The structure is NOT fully melted.

Reasoning

1. Visible long-range order: Throughout the box you can trace straight rows/planes of atoms that repeat periodically.

2. Clear lattice pattern: The atoms form a hexagonal/close-packed array with nearly uniform nearest-neighbour spacing.

3. Aligned packing: Atoms sit on well-defined lattice sites; only minor thermal vibrations or local defects are present.

4. Lack of uniform randomness: There are no large, completely disordered regions where the atomic positions lose their correlation.

Because crystalline order is still dominant, the sample has not reached the liquid state.

Recommended adjustments to reach complete melting

• Raise the target temperature (or broaden the temperature range).

• Extend the simulation time so the system can overcome kinetic barriers.

• Use a slower heating rate to avoid superheating and allow the lattice to break down gradually.

• Verify/adjust the interatomic potential parameters to be sure they reproduce the correct melting point.

Supplementary Fig. 16. Response of the o3 reasoning vision model on the image generated after the initial melting point calculation for iron with temperature range 1500→2500 K.

The structure is fully melted.

Reasoning

• Random distribution – The atoms fill the simulation box in a completely irregular way; there are no straight rows or planes that repeat through the volume.

• Absence of lattice structures – Nowhere in the image do parallel layers, stacked planes, or other periodic motifs appear.

• Non-aligned, close packing – Although the atoms are close to one another, each atom's nearest neighbours are oriented differently, not along common crystallographic directions.

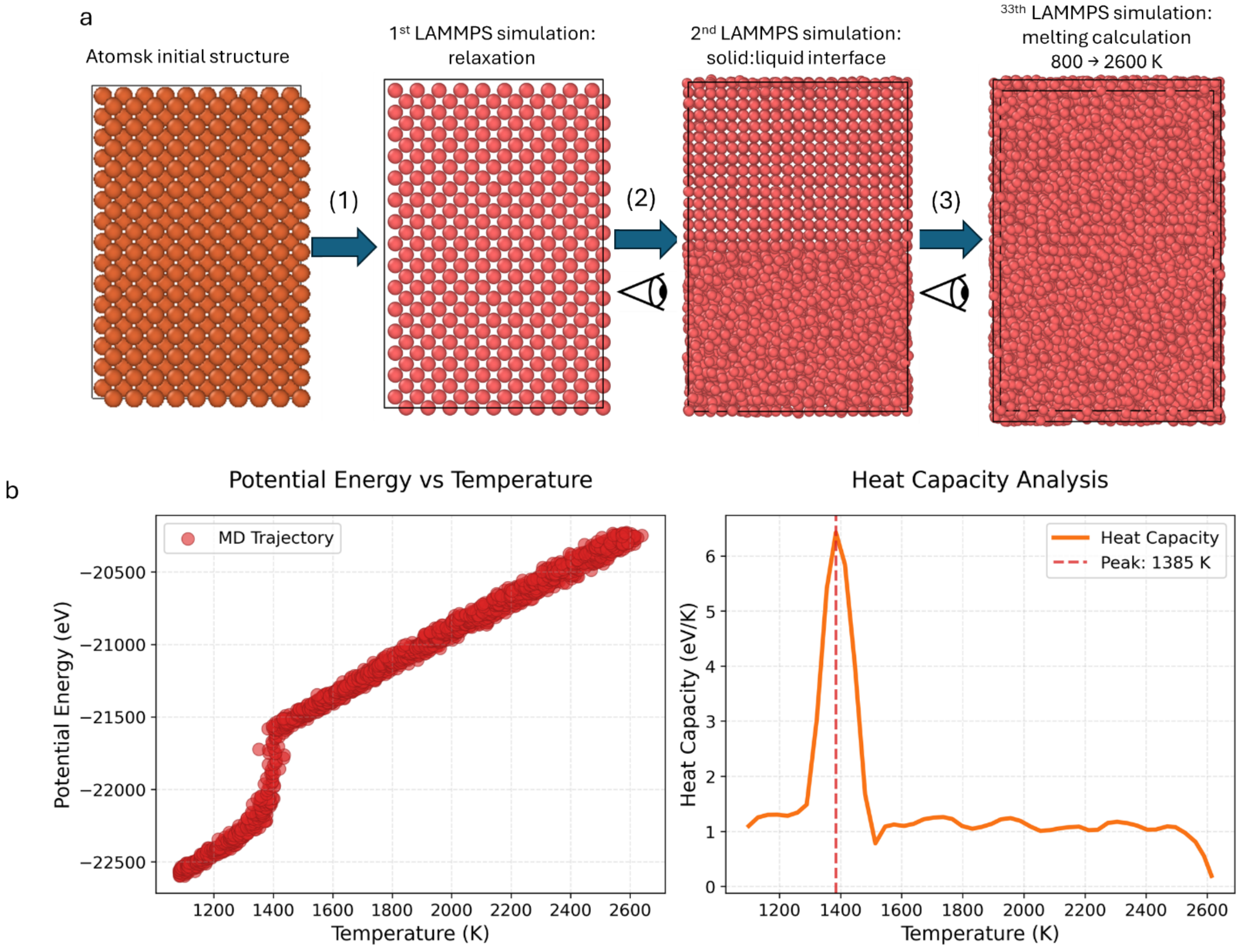
• Uniform disorder – The same disordered, fluid-like arrangement is seen from the centre out to the boundaries; there are no pockets of residual crystallinity or partially ordered grains.

Because these signatures match those of a liquid, no further parameter adjustments are required.

Supplementary Fig. 17. Response of the o3 reasoning vision model on the image generated after the initial melting point calculation for iron with temperature range 1500→3000 K.

2.4.1b Au (FCC)

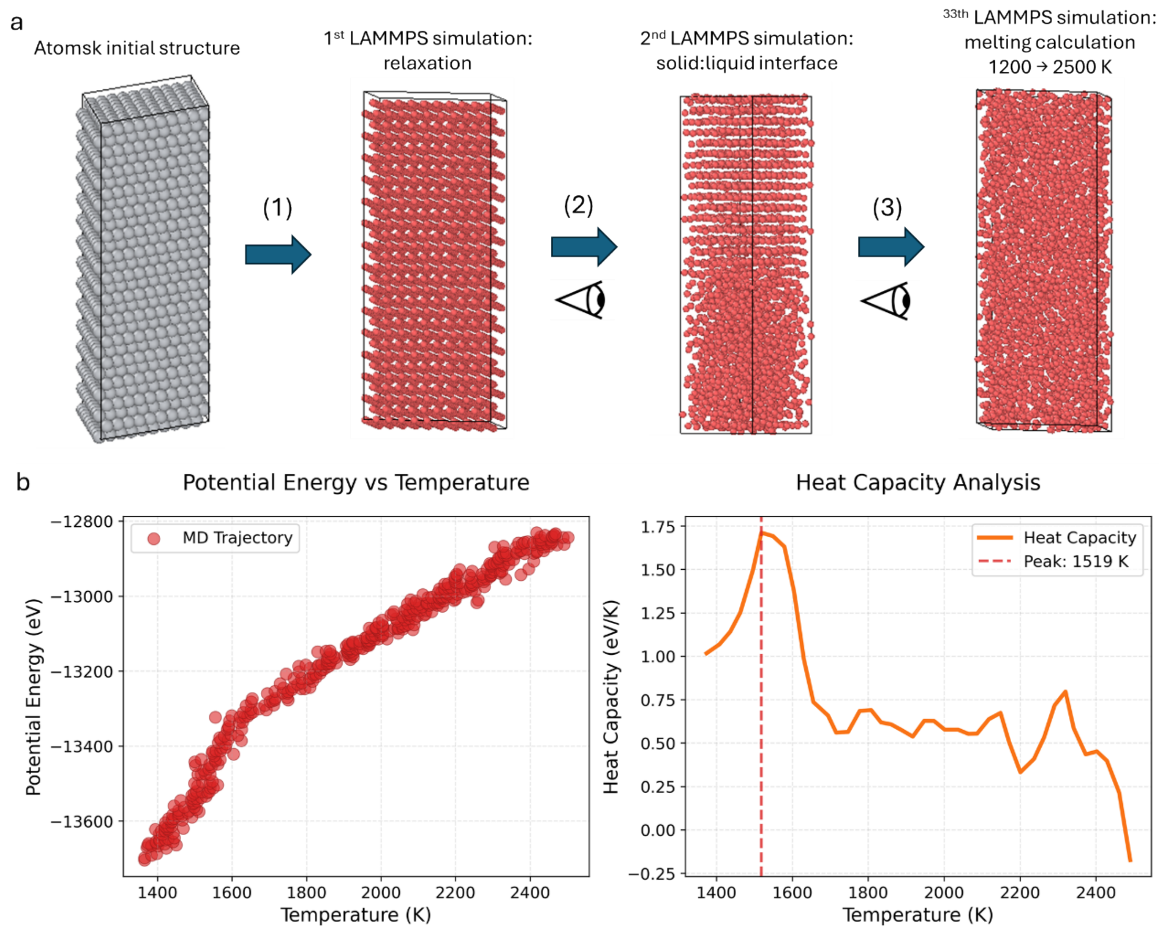
Task 4b: "Perform a melting point simulation for gold using LAMMPS."



Supplementary Fig. 18. a) Structure evolution during the progression of the melting point simulation executed autonomously by the agentic framework with integrated vision for the Au (FCC) system. The initial structure is generated by Atomsk and relaxed using LAMMPS. Then the relaxed structure is used as input to the 2nd LAMMPS simulation aiming to create a 50:50 solid liquid interface. Once this simulation terminates the analysis agent is creating an OVITO image from the last frame of the generated dump file. The interface image is given as an input to the vision agent to decide if it is 50:50 melted. Once the interface passes the assessment the structure is given as an input for the 3rd LAMMPS simulation which is the melting point. Once the simulation is finished and the last frame saved, the vision agents concludes that the structure was fully melted. b) Final generated report from the results analysis agent, displaying the calculated melting point as the peak in heat capacity.

2.4.1c Ti (HPC)

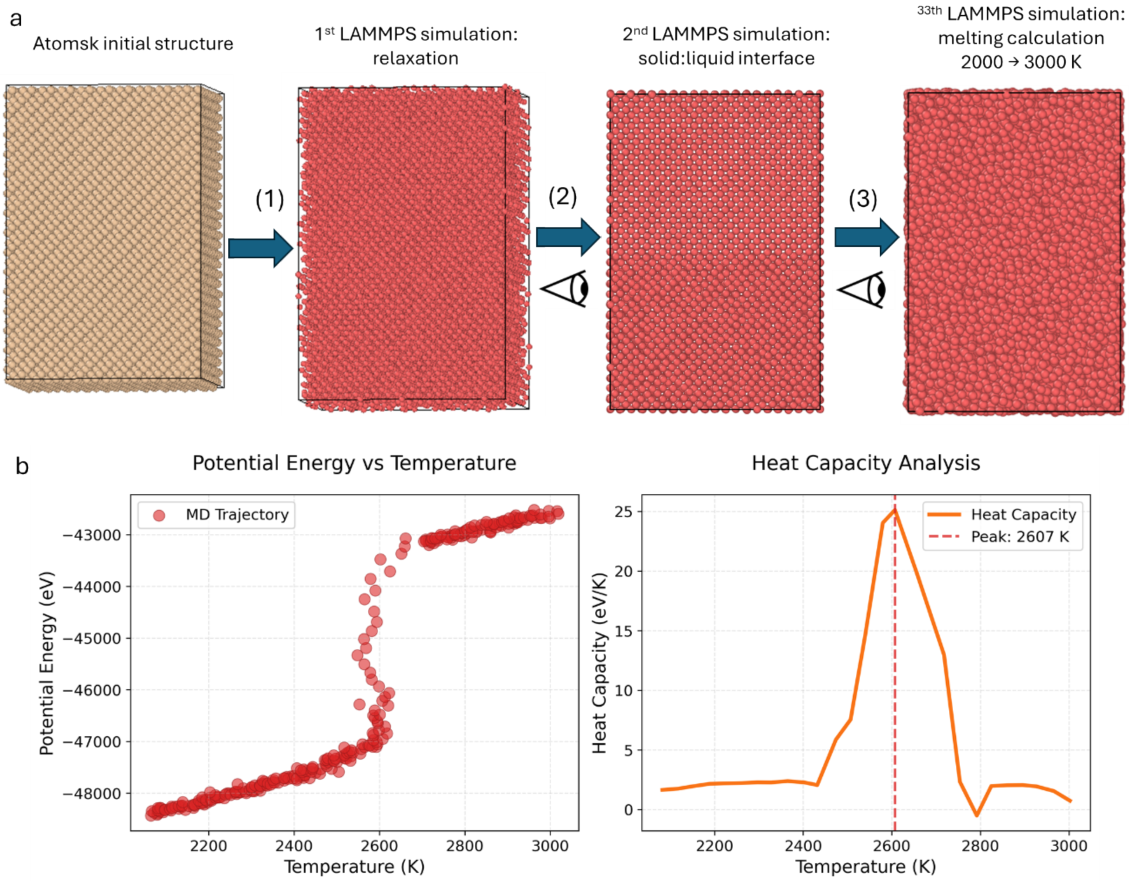
prompt_4c = """Perform a melting point simulation for a titanium using LAMMPS."""



Supplementary Fig. 19. a) Structure evolution during the progression of the melting point simulation executed autonomously by the agentic framework with integrated vision for the Ti (HPC) system. The initial structure is generated by Atomsk and relaxed using LAMMPS. Then the relaxed structure is used as input to the 2nd LAMMPS simulation aiming to create a 50:50 solid liquid interface. Once this simulation terminates the analysis agent is creating an OVITO image from the last frame of the generated dump file. The interface image is given as an input to the vision agent to decide if it is 50:50 melted. Once the interface passes the assessment the structure is given as an input for the 3rd LAMMPS simulation which is the melting point. Once the simulation is finished and the last frame saved, the vision agents concludes that the structure was fully melted. b) Final generated report from the results analysis agent, displaying the calculated melting point as the peak in heat capacity.

2.4.1c Si (Diamond)

prompt_4f = """Perform a melting point simulation for a gold-copper alloy using LAMMPS."""

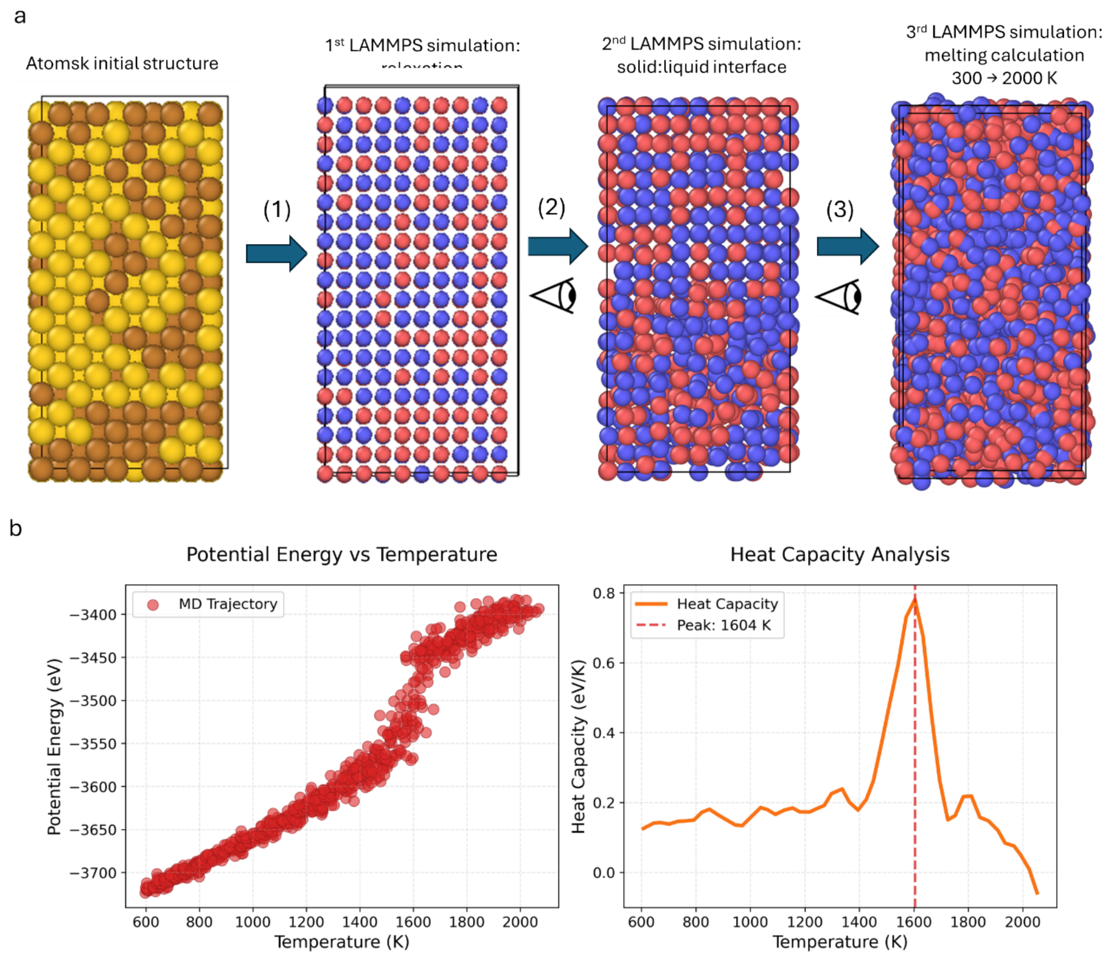


Supplementary Fig. 20. a) Structure evolution during the progression of the melting point simulation executed autonomously by the agentic framework with integrated vision for the silicon (diamond) system. The initial structure is generated by Atomsk and relaxed using LAMMPS. Then the relaxed structure is used as input to the 2nd LAMMPS simulation aiming to create a 50:50 solid liquid interface. Once this simulation terminates the analysis agent is creating an OVITO image from the last frame of the generated dump file. The interface image is given as an input to the vision agent to decide if it is 50:50 melted. Once the interface passes the assessment the structure is given as an input for the 3rd LAMMPS simulation which is the melting point. Once the simulation is finished and the last frame saved, the vision agents concludes that the structure was fully melted. b) Final generated report from the results analysis agent, displaying the calculated melting point as the peak in heat capacity.

2.4.2 Alloy systems

2.4.2a Au-Cu (FCC)

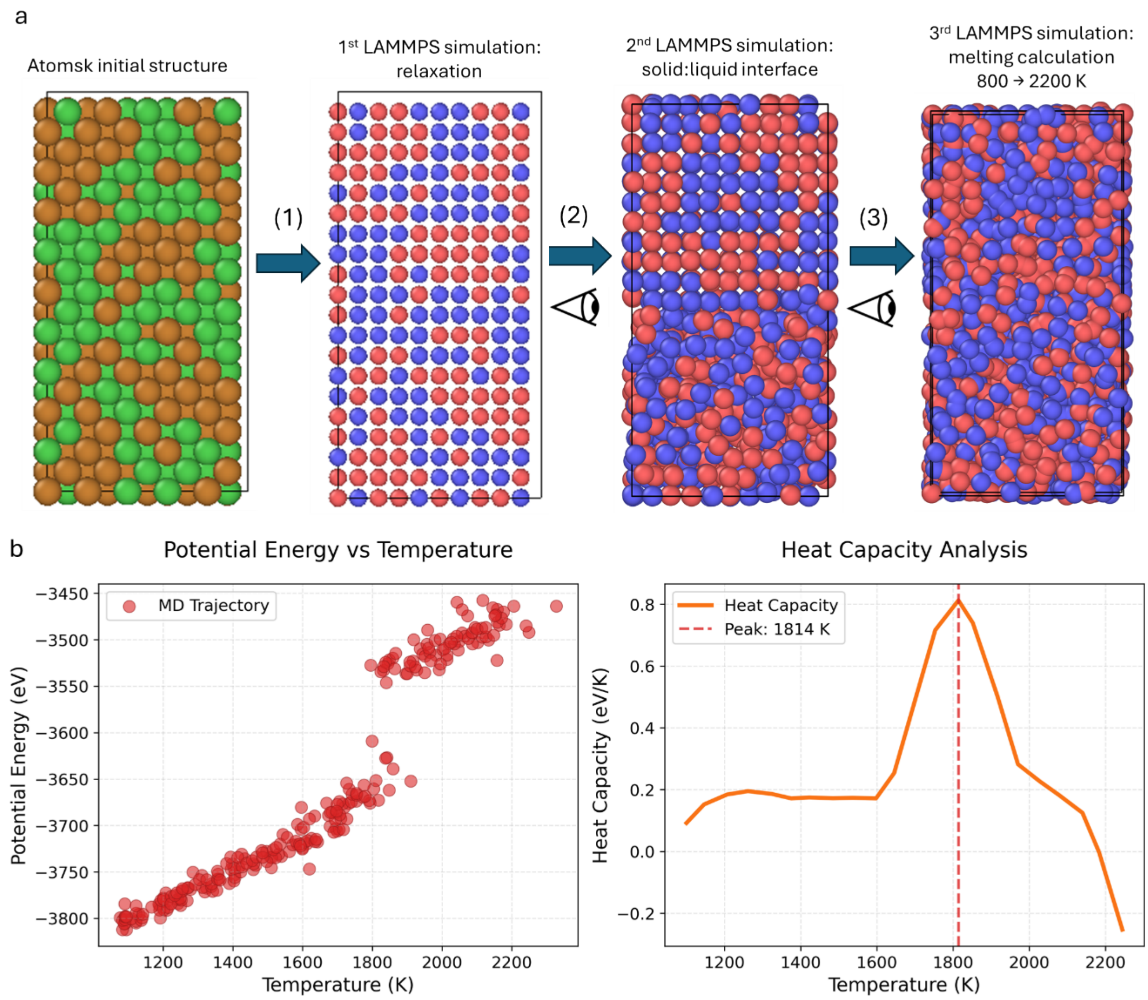
prompt_4e = """Perform a melting point simulation for a gold-copper alloy using LAMMPS."""



Supplementary Fig. 21. a) Structure evolution during the progression of the melting point simulation executed autonomously by the agentic framework with integrated vision for the gold-copper alloy (FCC) system. The initial structure is generated by Atomsk and relaxed using LAMMPS. Then the relaxed structure is used as input to the 2nd LAMMPS simulation aiming to create a 50:50 solid liquid interface. Once this simulation terminates the analysis agent is creating an OVITO image from the last frame of the generated dump file. The interface image is given as an input to the vision agent to decide if it is 50:50 melted. Once the interface passes the assessment the structure is given as an input for the 3rd LAMMPS simulation which is the melting point. Once the simulation is finished and the last frame saved, the vision agents concludes that the structure was fully melted. b) Final generated report from the results analysis agent, displaying the calculated melting point as the peak in heat capacity.

2.4.2b Ni-Cu (FCC)

prompt_4f = """Perform a melting point simulation for a nickel-copper alloy using LAMMPS."""



Supplementary Fig. 22. a) Structure evolution during the progression of the melting point simulation executed autonomously by the agentic framework with integrated vision for the nickel-copper alloy (FCC) system. The initial structure is generated by Atomsk and relaxed using LAMMPS. Then the relaxed structure is used as input to the 2nd LAMMPS simulation aiming to create a 50:50 solid liquid interface. Once this simulation terminates the analysis agent is creating an OVITO image from the last frame of the generated dump file. The interface image is given as an input to the vision agent to decide if it is 50:50 melted. Once the interface passes the assessment the structure is given as an input for the 3rd LAMMPS simulation which is the melting point. Once the simulation is finished and the last frame saved, the vision agents concludes that the structure was fully melted. b) Final generated report from the results analysis agent, displaying the calculated melting point as the peak in heat capacity.

Supplementary Table 5. Comparison of the calculated melting temperature (Tm) derived from the agentic system vs a human expert for the six diverse atomic systems investigated in this work. A literature reference is added for relevant property calculations. For the melting point, we note that quantitative comparison across studies is challenging because the predicted melting temperature is highly sensitive to simulation parameters such as heating rate, system size, and interatomic potential.

Crystal system	**Agent T_m**	**Human T_m**	**% error**	**Relevant literature reference**
Fe (BCC)	2765 K	2754 K	0.399%	[13]
Au (FCC)	1385 K	1416 K	2.10%	[14]
Ti (HPC)	1528 K	1533 K	0.326%	[14]
Si-Diamond	2602 K	2597 K	0.193%	[14]
Au-Cu (FCC)	1604 K	1597 K	0.438%	[15]
Ni-Cu (FCC)	1814 K	1830 K	0.874%	[16]

Supplementary Table 6. Summary table with the potential models (name and source) obtained by the PotentialManager/WebScraper as per crystal system and task.

Crystal system	**Task**	**Potential model**	**Reference**
Fe (BCC)	Task 1 Task 2 Task 3 Task 4	Finnis-Sinclair potential (LAMMPS cubic hermite tabulation) for bcc Fe developed by Ackland et al. (1997) v006 (https://openkim.org/id/EAM_Dynamo_AcklandBaconCalder_1997_Fe__MO_142799717516_006)	[17]
Au (FCC)	Task 1	EAM potential (LAMMPS cubic hermite tabulation) for Au developed by Ackland et al. (1987) v001(https://openkim.org/id/EAM_Dynamo_Ackland_1987_Au__MO_754413982908_001)	[18]
	Task 2 Task 3 Task 4	Au_u3.eam (https://raw.githubusercontent.com/lammps/lammps/develop/potentials/Au_u3.eam)	[19]
Ti (HPC)	Task 1 Task 2 Task 3 Task 4	Finnis-Sinclair potential (LAMMPS cubic hermite tabulation) for Ti for the hcp-fcc transition developed by Ackland (1992) v006 (https://openkim.org/id/EAM_Dynamo_Ackland_1992_Ti__MO_748534961139_006)	[20]
Si-Diamond	Task 1 Task 2 Task 3 Task 4	Stillinger-Weber potential for Si (https://raw.githubusercontent.com/lammps/lammps/develop/potentials/Si.sw)	[21]
Au-Cu (FCC)	Task 1 Task 2 Task 3 Task 4	EAM potential (LAMMPS cubic hermite tabulation) for Cu-Au alloys developed by Gola and Pastewka (2018) v001 (https://openkim.org/id/EAM_Dynamo_GolaPastewka_2018_CuAu__MO_426403318662_001)	[22]
Ni-Cu (FCC)	Task 1 Task 2 Task 3 Task 4	Cu-Ni EAM potential (DYNAMO setfl format) (https://openkim.org/id/EAM_Dynamo_OnatDurukanoglu_2014_CuNi__MO_592013496703_005)	[23]

References

1. Hirel, P. Atomsk: A tool for manipulating and converting atomic data files. *Comput Phys Commun* **197**, 212–219 (2015).

2. Atomsk. *https://github.com/pierrehirel/atomsk*.

3. Philipsen, P. H. T. & Baerends, E. J. Cohesive energy of 3d transition metals: Density functional theory atomic and bulk calculations. *Phys Rev B* **54**, 5326–5333 (1996).

4. Albritton, C. *Single Crystal Elastic Constants and Calculated Aggregate Properties*.

5. Epstein, S. G. & Carlson, O. N. The elastic constants of nickel-copper alloy single crystals. *Acta Metallurgica* **13**, 487–491 (1965).

6. Fisher, E. S. & Renken, C. J. Single-Crystal Elastic Moduli and the hcp \ensuremath{\rightarrow} bcc Transformation in Ti, Zr, and Hf. *Physical Review* **135**, A482–A494 (1964).

7. McSkimin, H. J. & Andreatch Jr., P. Elastic Moduli of Silicon vs Hydrostatic Pressure at 25.0°C and − 195.8°C. *J Appl Phys* **35**, 2161–2165 (1964).

8. Flinn, P. A., McManus, G. M. & Rayne, J. A. Elastic constants of ordered and disordered Cu3Au from 4.2 to 300°K. *Journal of Physics and Chemistry of Solids* **15**, 189–195 (1960).

9. Klotz, S. & Braden, M. Phonon Dispersion of bcc Iron to 10 GPa. *Phys Rev Lett* **85**, 3209–3212 (2000).

10. Lynn, J. W., Smith, H. G. & Nicklow, R. M. Lattice Dynamics of Gold. *Phys Rev B* **8**, 3493–3499 (1973).

11. Stassis, C., Arch, D., Harmon, B. N. & Wakabayashi, N. Lattice dynamics of hcp Ti. *Phys Rev B* **19**, 181–188 (1979).

12. Nilsson, G. & Nelin, G. Study of the Homology between Silicon and Germanium by Thermal-Neutron Spectrometry. *Phys Rev B* **6**, 3777–3786 (1972).

13. Royal Society of Chemistry, Periodic Table: Iron (Fe), https://www.rsc.org/periodic-table/element/26/iron , (accessed November 2025).

14. National Center for Biotechnology Information (NCBI), PubChem Periodic Table: Melting Points, https://pubchem.ncbi.nlm.nih.gov/ptable/melting-point/ , (accessed November 2025).

15. Chu, M. Z. *et al.* Melting and phase diagram of Au-Cu alloy at nanoscale. *J Alloys Compd* **891**, 162029 (2022).

16. Nickel Institute, Mechanical properties of copper-nickel alloys, https://nickelinstitute.org/en/nickel-applications/copper-nickel-alloys/mechanical-properties-of-copper-nickel-alloys/ , (accessed November 2025).

17. Ackland, G. J., Bacon, D. J., Calder, A. F. & Harry, T. Finnis-Sinclair potential (LAMMPS cubic hermite tabulation) for bcc Fe developed by Ackland et al. (1997) v006. Preprint at https://doi.org/10.25950/e26dba93 (2025).

18. Ackland, G. J., Tichy, G., Vitek, V. & M.W., F. Simple N-body potentials for the noble-metals and nickel. *Phil. Mag. A - Phys. Cond. Matter* **56**, 735–756 (1987).

19. Foiles, S. M., Baskes, M. I. & Daw, M. S. Embedded-atom-method functions for the fcc metals Cu, Ag, Au, Ni, Pd, Pt, and their alloys. *Phys Rev B* **33**, 7983–7991 (1986).

20. Ackland, G. J. Theoretical study of titanium surfaces and defects with a new many-body potential. *Philosophical Magazine A* **66**, 917–932 (1992).

21. Stillinger, F. H. & Weber, T. A. Computer simulation of local order in condensed phases of silicon. *Phys Rev B* **31**, 5262–5271 (1985).

22. Grochola, G., Russo, S. P. & Snook, I. K. On fitting a gold embedded atom method potential using the force matching method. *J Chem Phys* **123**, 204719 (2005).

23. Onat, B. & Durukanoğlu, S. An optimized interatomic potential for Cu–Ni alloys with the embedded-atom method. *Journal of Physics: Condensed Matter* **26**, 035404 (2014).