Supporting Information

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Machine Learning to Predict Quasicrystals from Chemical Compositions

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Supplementary Note

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Table S1: List of quasicrystals and approximants that were used in the supervised learning. Each column gives the chemical composition, the label indicating quasicrystal or approximant (QC and AC), structure type, the known space group, and the guessed space group, respectively. The data were collected from the Crystallography of Quasicrystals handbook [1].

| Composition          | QC or AP | Type      | Space group   | Guessed space group |
|----------------------|----------|-----------|---------------|---------------------|
| Cr 5 Ni 3 Si 2       | QC       | Octagonal | I8_4/mcm      |                     |
| Mn 4 Si 1            | QC       | Octagonal | I8_4/mcm      |                     |
| Mn 82 Si 15 Al 3     | QC       | Octagonal | I8_4/mcm      | I8_4/mcm            |
| Zn 58 Mg 40 Dy 2     | QC       | Decagonal | P10_5/mmc     |                     |
| Zn 58 Mg 40 Dy 2     | QC       | Decagonal | P10_5/mmc     |                     |
| Zn 58 Mg 40 Er 2     | QC       | Decagonal | P10_5/mmc     |                     |
| Zn 58 Mg 40 Ho 2     | QC       | Decagonal | P10_5/mmc     |                     |
| Zn 58 Mg 40 Lu 2     | QC       | Decagonal | P10_5/mmc     |                     |
| Zn 58 Mg 40 Tm 2     | QC       | Decagonal | P10_5/mmc     |                     |
| Zn 58 Mg 40 Y 2      | QC       | Decagonal | P10_5/mmc     |                     |
| Al 70 Ni 15 Fe 15    | QC       | Decagonal | P10_5/mmc     |                     |
| Al 65 Cu 15 Rh 20    | QC       | Decagonal | P10_5/mmc     |                     |
| Al 70 Ni 20 Rh 10    | QC       | Decagonal | P10_5/mmc     |                     |
| Al 65 Cu 20 Co 15    | QC       | Decagonal | P10_5/mmc     |                     |
| Al 65 Cu 20 Ir 15    | QC       | Decagonal | P10_5/mmc     |                     |
| Al 73.2 Co 26.8      | AP       | Decagonal | P6_3/mmc      | P6_3/mmc            |
| Al 71.5 Co 28.5      | AP       | Decagonal | Cm            | Cm                  |
| Al 71.5 Co 23.6 Cu 4.9 | AP   | Decagonal | Pmm2_1         | Pmm2_1              |
| Al 76 Co 24          | AP       | Decagonal | Immm          | Immm                |
| Al 71.8 Co 21.1 Ni 7.1 | AP   | Decagonal | P10_5/mmc     |                     |
| Al 70.5 Mn 16.5 Pd 13 | QC   | Decagonal | P10_5/mmc     |                     |
| Al 40 Mn 25 Fe 15 Ge 20 | QC | Decagonal | P10_5/mmc     |                     |
| Ga 33 Fe 46 Cu 3 Si 18 | QC  | Decagonal | P10_5/mmc     |                     |
| Ga 43 Co 47 Cu 10    | QC       | Decagonal | P10_5/mmc     |                     |
| Ga 35 V 45 Ni 6 Si 14 | QC   | Decagonal | P10_5/mmc     |                     |
| Al 75 Mn 20 Pd 5     | AP       | Decagonal | Pnma          |                     |
| Al 75 Os 10 Pd 15    | QC       | Decagonal | P10_5/mmc     |                     |
| Al 75 Ru 10 Pd 15    | QC       | Decagonal | P10_5/mmc     |                     |
| Al 73 Ir 14.5 Os 12.5 | QC | Decagonal | P10_c          | P10_c               |
| Al 70 Ni 20 Ru 10    | QC       | Decagonal | P10_5/mmc     |                     |
| Al 72.5 Pd 27.5      | AP       | Decagonal | Pna2_1         | Pna2_1              |
| Al 75 Pd 13 Ru 12    | AP       | Decagonal | Pna2_1         | Pna2_1              |
| Ni 70.6 Cr 29.4      | QC       | Dodecagonal | (-12)m2       |                     |
| V 3 Ni 2             | QC       | Dodecagonal | (-12)m2       |                     |
| V 15 Ni 10 Si 1      | QC       | Dodecagonal | (-12)m2       |                     |
| Ta 1.6 Te 1          | QC       | Dodecagonal | (-12)m2       |                     |
| Ta 97 Te 60          | AP       | Dodecagonal | P2_12_12_1    | P2_12_12_1          |
| Ta 181 Te 112        | AP       | Dodecagonal | P2_12_12_1    | P2_12_12_1          |
| Al 65 Cu 20 Ru 15    | QC       | Icosahedral | Fm-3-5        | Fm-3-5              |
| Al 65 Cu 20 Os 15    | QC       | Icosahedral | Fm-3-5        | Fm-3-5              |
| Al 65 Cu 20 Fe 15    | QC       | Icosahedral | Fm-3-5        | Fm-3-5              |
| Composition       | QC or AP | Type         | Space group | Guessed space group |
|-------------------|----------|--------------|-------------|---------------------|
| Al 70.5 Pd 21 Mn 8.5 | QC       | Icosahedral  | Fm-3-5      | Fm-3-5              |
| Al 70 Pd 20 Re 10  | QC       | Icosahedral  | Fm-3-5      | Fm-3-5              |
| Al 70 Pd 21 Tc 9   | QC       | Icosahedral  | Fm-3-5      | Fm-3-5              |
| Al 71 Pd 21 Re 8   | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Al 72 Pd 17 Ru 11  | QC       | Icosahedral  | Fm-3-5      | Fm-3-5              |
| Al 72 Pd 17 Os 11  | QC       | Icosahedral  | Fm-3-5      | Fm-3-5              |
| Al 71.5 Cu 8.5 Ru 20 | AP 1/0  | Icosahedral  | P2_13       | P2_13               |
| Al 55.1 Cu 14.6 Ru 20.2 Si 10.1 | AP 1/0×2 (γ) | Icosahedral  | Fm-3       | Fm-3               |
| Al 39 Pd 21 Fe 2  | AP 1/0×2 (c) | Icosahedral  | Fm-3       | Fm-3               |
| Al 68 Pd 20 Ru 12  | AP 1/0×2 (c) | Icosahedral  | P23        | P23                |
| Al 57.3 Cu 31.4 Ru 11.3 | AP 1/1  | Icosahedral  | Pm-3       | Pm-3               |
| Al 55 Cu 25.5 Fe 12.5 Si 7 | AP 1/1 | Icosahedral  | Pm-3       | Pm-3               |
| Al 40 Mn 10.1 Si 7.4 | AP 1/1  | Icosahedral  | Pm-3       | Pm-3               |
| Al 67 Pd 11 Mn 14 Si 7 | AP 1/1  | Icosahedral  | Pm-3       | Pm-3               |
| Al 66.6 Rh 26.1 Si 7.3 | AP 2/1  | Icosahedral  | Pm-3       | Pm-3               |
| Al 70 Pd 23 Mn 6 Si 1 | AP 2/1  | Icosahedral  | Pm-3       | Pm-3               |
| Zn 76 Mg 17 Hf 7   | QC       | Icosahedral  | Fm-3-5      | Fm-3-5              |
| Zn 84 Mg 7 Zr 9    | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Al 6 Cu 1 Li 3     | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Mg 43 Al 42 Pd 15  | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Zn 40 Mg 39.5 Ga 25 | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Zn 74 Mg 15 Ho 11  | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Zn 56.8 Mg 34.6 Tb 8.7 | QC     | Icosahedral  | Fm-3-5      | Fm-3-5              |
| Zn 65 Mg 26 Ho 9   | QC       | Icosahedral  | Fm-3-5      | Fm-3-5              |
| Zn 64 Mg 25 Y 11   | QC       | Icosahedral  | Fm-3-5      | Fm-3-5              |
| Zn 55 Mg 40 Nd 5   | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Zn 56.8 Mg 34.6 Dy 8.7 | QC     | Icosahedral  | Fm-3-5      | Fm-3-5              |
| Zn 77 Mg 17.5 Ti 5.5 | AP 1/1  | Icosahedral  | Pm-3       | Pm-3               |
| Zn 77 Mg 18 Hf 5   | AP 1/1  | Icosahedral  | Pm-3       | Pm-3               |
| Zn 77 Mg 18 Zr 5   | AP 1/1  | Icosahedral  | Pm-3       | Pm-3               |
| Al 88.6 Cu 19.4 Li 50.3 | AP 1/1 | Icosahedral  | Im-3       | Im-3               |
| Zn 34.6 Mg 40 Al 25.4 | AP 1/1  | Icosahedral  | Im-3       | Im-3               |
| Zn 61.4 Mg 24.5 Er 14.1 | AP 2/1  | Icosahedral  | F-43m      | F-43m              |
| Zn 37 Mg 46 Al 17  | AP 2/1  | Icosahedral  | Pm-3       | Pm-3               |
| Zn 47.3 Mg 27 Al 10.7 | AP 2/1  | Icosahedral  | Pa-3       | Pa-3               |
| Zn 73.6 Mg 2.5 Sc 11.2 | AP 2/1  | Icosahedral  | Pa-3       | Pa-3               |
| Zn 40 Mg 39.5 Ga 16.4 Al 4.1 | AP 3/2-2/1-2/1 | Icosahedral  | Cmc2_1     | Cmc2_1             |
| Cu 48 Sc 15 Ga 34 Mg 3   | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Cu 46 Sc 16 Al 38      | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Zn 84 Ti 8 Mg 8       | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Zn 75 Sc 15 Ni 10     | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Zn 72 Sc 16 Cu 12     | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Zn 75 Sc 15 Co 10     | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Zn 75 Sc 15 Fe 10     | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Zn 75 Sc 15 Mn 10     | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Zn 80 Sc 15 Mg 5      | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Zn 75 Sc 15 Pt 10     | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Zn 75 Sc 15 Pd 10     | QC       | Icosahedral  | Pm-3-5      | Pm-3-5              |
| Composition       | QC or AP | Type         | Space group | Guessed space group |
|-------------------|----------|--------------|-------------|--------------------|
| Zn 75 Sc 15 Au 10 | QC       | Icosahedral  | Pm-3-5      |                    |
| Zn 75 Sc 15 Ag 10 | QC       | Icosahedral  | Pm-3-5      |                    |
| Zn 77 Sc 8 Ho 8 Fe 7 | QC       | Icosahedral  | Pm-3-5      |                    |
| Zn 77 Sc 8 Er 8 Fe 7 | QC       | Icosahedral  | Pm-3-5      |                    |
| Zn 77 Sc 7 Tm 9 Fe 7 | QC       | Icosahedral  | Pm-3-5      |                    |
| Zn 56.8 Er 8.7 Mg 34.6 | QC       | Icosahedral  | Fm-3-5      |                    |
| Zn 76 Yb 14 Mg 10 | QC       | Icosahedral  | Fm-3-5      |                    |
| Cd 65 Mg 20 Lu 15 | QC       | Icosahedral  | Pm-3-5      |                    |
| Ag 42 In 42 Yb 16 | QC       | Icosahedral  | Pm-3-5      |                    |
| Cd 65 Mg 20 Tm 15 | QC       | Icosahedral  | Pm-3-5      |                    |
| Ag 42 In 42 Ca 16 | QC       | Icosahedral  | Pm-3-5      |                    |
| Au 44.2 In 41.7 Ca 14.1 | QC       | Icosahedral  | Pm-3-5      |                    |
| Cd 65 Mg 20 Y 15 | QC       | Icosahedral  | Pm-3-5      |                    |
| Cd 65 Mg 20 Er 15 | QC       | Icosahedral  | Fm-3-5      |                    |
| Cd 65 Mg 20 Ho 15 | QC       | Icosahedral  | Fm-3-5      |                    |
| Cd 65 Mg 20 Tb 15 | QC       | Icosahedral  | Fm-3-5      |                    |
| Cd 65 Mg 20 Dy 15 | QC       | Icosahedral  | Fm-3-5      |                    |
| Cd 65 Mg 20 Gd 15 | QC       | Icosahedral  | Fm-3-5      |                    |
| Cd 84 Yb 16       | QC       | Icosahedral  | Pm-3-5      |                    |
| Cd 65 Mg 20 Yb 15 | QC       | Icosahedral  | Pm-3-5      |                    |
| Cd 85 Ca 15       | QC       | Icosahedral  | Pm-3-5      |                    |
| Cd 65 Mg 20 Ca 15 | QC       | Icosahedral  | Pm-3-5      |                    |
| Be 17 Ru 3        | AP 1/1   | Icosahedral  | Im-3        |                    |
| Ga 3.85 Ni 2.15 Hf 1 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Ga 3.22 Ni 2.78 Zr 1 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Ga 3.64 Ni 2.36 Sc 1 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Ga 2.3 Cu 3.7 Sc 1 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Ga 2.6 Cu 3.4 Lu 1 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Zn 17 Sc 3        | AP 1/1   | Icosahedral  | Im-3        |                    |
| Zn 17 Yb 3        | AP 1/1   | Icosahedral  | Im-3        |                    |
| Ag 42.5 Ga 42.5 Yb 15 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Au 50.5 Ga 35.9 Ca 13.6 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Au 61.2 Sn 23.9 Dy 15.2 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Au 62.3 Sn 23.1 Gd 14.6 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Ag 42.2 In 42.6 Tm 15.2 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Au 47.2 In 37.2 Gd 15.6 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Au 12.2 In 6.3 Ca 3 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Au 64.2 Sn 21.3 Pr 14.5 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Au 65 Sn 20 Ce 15  | AP 1/1   | Icosahedral  | Im-3        |                    |
| Ag 46.4 In 39.7 Gd 13.9 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Au 49.7 In 35.4 Ce 14.9 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Au 60.7 Sn 25.2 Eu 14.1 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Ag 2 In 4 Yb 1    | AP 1/1   | Icosahedral  | Im-3        |                    |
| Ag 46.9 In 38.7 Pr 14.4 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Au 42 In 42 Yb 16 | AP 1/1   | Icosahedral  | Ia-3        |                    |
| Cd 6 Gd 1         | AP 1/1   | Icosahedral  | Im-3        |                    |
| Ag 2 In 4 Ca 1    | AP 1/1   | Icosahedral  | Im-3        |                    |
| Ag 47.7 In 38.7 Ce 14.2 | AP 1/1   | Icosahedral  | Im-3        |                    |
| Composition | QC or AP | Type     | Space group | Guessed space group |
|-------------|----------|----------|-------------|---------------------|
| Cd 6 Dy 1   | AP 1/1   | Icosahedral | Im-3        | Im-3                |
| Cd 6 Y 1    | AP 1/1   | Icosahedral | Im-3        | Im-3                |
| Cd 6 Sm 1   | AP 1/1   | Icosahedral | Im-3        | Im-3                |
| Cd 6 Nd 1   | AP 1/1   | Icosahedral | Im-3        | Im-3                |
| Cd 6 Yb 1   | AP 1/1   | Icosahedral | Im-3        | Im-3                |
| Ag 42.9 In 43.6 Eu 13.5 | AP 1/1 | Icosahedral | Im-3        | Im-3                |
| Cd 6 Ca 1   | AP 1/1   | Icosahedral | Im-3        | Im-3                |
| Cd 25 Eu 4  | AP 1/1×2 | Icosahedral | Fd-3        | Fd-3                |
| Cd 19 Pr 3  | AP 1/1   | Icosahedral | Im-3        | Im-3                |
| Cd 6 Sr 1   | AP 1/1   | Icosahedral | Im-3        | Im-3                |
| Au 61.1 Ga 25.0 Ca 13.9 | AP 2/1 | Icosahedral | Pa-3        | Pa-3                |
| Au 60.3 Sn 24.6 Yb 15.1 | AP 2/1 | Icosahedral | Pa-3        | Pa-3                |
| Au 61.2 Sn 24.3 Ca 14.5 | AP 2/1 | Icosahedral | Pa-3        | Pa-3                |
| Au 42.9 In 41.9 Yb 15.2 | AP 2/1 | Icosahedral | Pa-3        | Pa-3                |
| Au 37 In 39.6 Ca 12.6 | AP 2/1 | Icosahedral | Pa-3        | Pa-3                |
| Ag 41.7 In 43.2 Yb 15.1 | AP 2/1 | Icosahedral | Pa-3        | Pa-3                |
| Au 61.2 Sn 24.5 Eu 14.3 | AP 2/1 | Icosahedral | Pa-3        | Pa-3                |
| Ag 41 In 44 Yb 15 | AP 2/1 | Icosahedral | Pa-3        | Pa-3                |
| Ag 42 In 45 Ca 13 | AP 2/1 | Icosahedral | Pa-3        | Pa-3                |
| Cd 76 Ca 13 | AP 2/1   | Icosahedral | Pa-3        | Pa-3                |
| Ag 43.4 In 42.8 Eu 13.8 | AP 2/1 | Icosahedral | Pa-3        | Pa-3                |
**Element features**

Table S2: 58 element features that were used to calculate the compositional descriptors. The dataset is accessible at our open-source software XenonPy [2].

| Feature ID                  | Description                                           | Unit                  | Reference |
|-----------------------------|-------------------------------------------------------|-----------------------|-----------|
| atomic_number               | Atomic number                                         |                       | [3, 4]    |
| atomic_radius               | Atomic radius                                          | pm                    |           |
| atomic_radius_rahm          | Atomic radius by Rahm et al.                          | pm                    | [4–6]     |
| atomic_volume               | Atomic volume                                          | cm$^3$ mol$^{-1}$     | [4]       |
| atomic_weight               | Atomic weight                                          |                       | [4, 7, 8] |
| boiling_point               | Boiling temperature                                    | K                     | [4]       |
| bulk_modulus                | Bulk modulus                                           | GPa                   | [9]       |
| c6_gb                       | C$_6$ dispersion coefficient in a.u.                  | a.u.                  | [4, 10, 11] |
| covalent_radius_cordero     | Covalent radius by Cerdero et al.                     | pm                    | [4, 12]   |
| covalent_radius_pyykko      | Single bond covalent radius by Pyykko et al.          | pm                    | [4, 13]   |
| covalent_radius_pyykko_double | Double bond covalent radius by Pyykko et al. | pm                    | [4, 14]   |
| covalent_radius_pyykko_triple | Triple bond covalent radius by Pyykko et al.          | pm                    | [4, 15]   |
| covalent_radius_slater      | Covalent radius by Slater                             | pm                    | [3]       |
| density                     | Density at 295K                                       | g cm$^3$              | [4]       |
| dipole_polarizability       | Dipole polarizability                                 | a.u.,                 | [4, 16]   |
| electron_negativity         | Pauling electronegativity                             |                       | [17]      |
| electron_affinity           | Electron affinity                                      | eV                    | [4, 18, 19] |
| en_allen                    | Allen’s scale of electronegativity                     | eV                    | [4, 20, 21] |
| en_ghosh                    | Ghosh’s scale of electronegativity                     |                       | [4, 22]   |
| en_pauling                  | Pauling’s scale of electronegativity                   |                       | [4, 18]   |
| first_ion_en                | First ionization energy                               | eV                    | [18]      |
| fusion_enthalpy             | Enthalpy of fusion for elements at their melting       | kJ mol$^{-1}$         | [18]      |
| gs_bandgap                  | DFT bandgap energy of T=0K ground state                | eV                    | [23, 24]  |
| gs_energy                   | DFT energy per atom (raw VASP value) of T=0K ground    | eV atom$^{-1}$        | [23, 24]  |
| gs_est_bcc_latcnt           | Estimated BCC lattice parameter based on the DFT      |                       | [23, 24]  |
|                            | volume of the OQMD ground state for each element       |                       |           |
| gs_est_fcc_latcnt           | Estimated FCC lattice parameter based on the DFT      |                          | [23, 24]  |
|                            | volume of the OQMD ground state for each element       |                          |           |
| gs_mag_moment               | DFT magnetic moment of T=0K ground state               |                      | [23, 24]  |
| gs_volume_per               | DFT volume per atom of T=0K ground state               | Å$^3$ atom$^{-1}$     | [23, 24]  |
| hhi_p                       | HerfindahlHirschman Index (HHI) production values      |                       | [25]      |
| hhi_r                       | HerfindahlHirschman Index (HHI) reserves values        |                       | [25]      |
| Feature ID               | Description                                                                 | Unit                           | Reference |
|-------------------------|-----------------------------------------------------------------------------|--------------------------------|-----------|
| heat_capacity_mass      | Specific heat capacity at STP\(^1\)                                         | J mol\(^{-1}\) K\(^{-1}\)     | [18]      |
| heat_capacity_molar     | Molar heat capacity at STP                                                  | J mol\(^{-1}\) K\(^{-1}\)     | [18]      |
| icsd_volume             | Volume per atom of ICSD phae at STP                                         |                                | [26–28]   |
| evaporation_heat        | Evaporation heat                                                            | kJ mol\(^{-1}\)               | [4]       |
| heat_offormation        | Heat of formation                                                           | kJ mol\(^{-1}\)               | [4]       |
| lattice_constant        | Lattice constant                                                            | Å                               | [4]       |
| mendeleev_number        | Mendeleev’s number                                                          |                                 | [4, 29, 30]|
| melting_point           | Melting temperature                                                         | K                               | [4]       |
| molar_volume            | Molar volume                                                               | L mol\(^{-1}\)                | [9]       |
| num_unfilled            | Number of unfilled valence orbitals                                        |                                | [31, 32]  |
| num_valance             | Number of valence electrons                                                |                                | [31, 32]  |
| num_d_unfilled          | Number of unfilled d valence orbitals                                      |                                | [31, 32]  |
| num_d_valence           | Number of filled d valence orbitals                                        |                                | [31, 32]  |
| num_f_unfilled          | Number of unfilled f valence orbitals                                      |                                | [31, 32]  |
| num_f_valence           | Number of filled f valence orbitals                                        |                                | [31, 32]  |
| num_s_unfilled          | Number of unfilled s valence orbitals                                      |                                | [31, 32]  |
| num_s_valence           | Number of filled s valence orbitals                                        |                                | [31, 32]  |
| period                  | Period in periodic table                                                    |                                | [4]       |
| specific_heat           | Specific heat at 20 °C                                                      | J g\(^{-1}\) mol\(^{-1}\)     | [4]       |
| thermal_conductivity    | Thermal conductivity at 25 °C                                                | W m\(^{-1}\) K\(^{-1}\)       | [4]       |
| vdw_radius              | van der Waals radius                                                        | pm                             | [4, 18]   |
| vdw_radius_alvarez      | van der Waals radius according to Alvarez                                   | pm                             | [4, 33]   |
| vdw_radius_mm3          | van der Waals radius from the MM3 FF                                        | pm                             | [4, 34]   |
| vdw_radius_uff          | van der Waals radius from the UFF                                            | pm                             | [4, 35]   |
| sound_velocity          | Velocity of sound                                                           | m s\(^{-1}\)                  | [9]       |
| polarizability          | Static average electric dipole polarizability                               | \(10^{-24}\) cm\(^3\)         | [18]      |

\(^1\)Standard Temperature and Pressure
Generalization ability of the model

Table S3: Prediction performance for the three-class classification problem of stable quasicrystals (QC), approximants (AC), and others. The left table is the confusion matrix, and the right table reports the per-class recall, precision, and $F_1$ metrics. The performance metrics were averaged over 100 different bootstrap sets, and the numbers in parentheses represent the standard deviations. The results labeled as “with Al systems” are the same as we report in Table 1. Additionally, in the “without Al systems” panel, we show the results of the performance evaluation after eliminating the aluminum-containing compositions from the test instances.

| True class | Test set | QC     | AC     | others |
|------------|----------|--------|--------|--------|
| With Al systems | QC       | 9.63 (1.641) | 3.24 (1.342) | 3.13 (1.189) |
|             | AC       | 3.11 (1.555) | 9.73 (1.805) | 3.16 (1.573) |
|             | others   | 0.76 (0.896) | 0.42 (0.619) | 2016.82 (1.024) |
| Without Al systems | QC       | 7.14 (1.594) | 1.91 (1.150) | 1.90 (1.221) |
|                 | AC       | 1.43 (1.185) | 7.68 (1.788) | 2.36 (1.389) |
|                 | others   | 0.45 (0.698) | 0.16 (0.367) | 1900.13 (8.795) |

| Predicted class | Performance metrics |
|-----------------|---------------------|
|                 | Recall | Precision | $F_1$ |
| QC              | 0.602 (0.103) | 0.722 (0.090) | 0.650 (0.076) |
| AC              | 0.608 (0.113) | 0.731 (0.089) | 0.658 (0.088) |
| others          | 0.999 (0.001) | 0.997 (0.001) | 0.998 (0.001) |
| QC              | 0.662 (0.140) | 0.807 (0.125) | 0.715 (0.102) |
| AC              | 0.671 (0.137) | 0.792 (0.110) | 0.720 (0.108) |
| others          | 1.000 (0.000) | 0.998 (0.001) | 0.999 (0.001) |
Table S4: The sensitivity of the prediction accuracy when using the different sample sizes of periodic crystals ($N_{\text{crystal}}$). The left table is the confusion matrix, and the right table reports the per-class recall, precision, and F1 metrics. The performance metrics were averaged over 100 different bootstrap sets, and the numbers in parentheses represent the standard deviations.

| $N_{\text{crystal}}$ | Predicted class | Performance metrics |
|----------------------|----------------|---------------------|
|                      | QC         | AC       | others | Recall    | Precision | F1       |
| 500                  | QC         | 11.08 (2.58) | 3.58 (1.97) | 0.88 (1.04) | 0.719 (0.121) | 0.736 (0.107) | 0.719 (0.086) |
|                      | AC         | 3.48 (1.75)  | 10.75 (2.98) | 1.13 (1.12)  | 0.701 (0.125) | 0.735 (0.127) | 0.708 (0.096) |
|                      | others     | 0.61 (0.84)  | 0.35 (0.64)  | 118.14 (4.25)| 0.992 (0.009) | 0.983 (0.012) | 0.988 (0.007) |
| 2,500                | QC         | 9.90 (3.10)  | 3.65 (1.80)  | 2.19 (1.44)  | 0.627 (0.132) | 0.718 (0.120) | 0.661 (0.109) |
|                      | AC         | 3.48 (1.82)  | 9.88 (2.59)  | 2.03 (1.38)  | 0.645 (0.120) | 0.708 (0.107) | 0.668 (0.093) |
|                      | others     | 0.38 (0.61)  | 0.49 (0.67)  | 518.00 (4.82)| 0.998 (0.002) | 0.992 (0.004) | 0.995 (0.002) |
| 5,000                | QC         | 9.96 (2.83)  | 3.47 (1.77)  | 2.40 (1.41)  | 0.627 (0.123) | 0.756 (0.128) | 0.677 (0.102) |
|                      | AC         | 2.95 (1.81)  | 9.67 (2.75)  | 2.84 (1.68)  | 0.631 (0.124) | 0.704 (0.120) | 0.655 (0.096) |
|                      | others     | 0.37 (0.67)  | 0.66 (0.78)  | 1017.68 (4.86)| 0.999 (0.001) | 0.995 (0.002) | 0.997 (0.001) |
| 10,000               | QC         | 9.73 (2.75)  | 3.17 (1.62)  | 3.02 (1.68)  | 0.609 (0.135) | 0.758 (0.127) | 0.666 (0.112) |
|                      | AC         | 2.68 (1.70)  | 8.54 (2.33)  | 3.67 (1.98)  | 0.584 (0.127) | 0.694 (0.112) | 0.623 (0.091) |
|                      | others     | 0.50 (0.70)  | 0.72 (0.83)  | 2017.97 (5.19)| 0.999 (0.001) | 0.997 (0.001) | 0.998 (0.001) |
| 15,000               | QC         | 9.47 (2.94)  | 3.33 (2.04)  | 3.49 (1.58)  | 0.582 (0.133) | 0.736 (0.113) | 0.641 (0.105) |
|                      | AC         | 2.90 (1.66)  | 8.46 (2.22)  | 4.65 (2.44)  | 0.536 (0.123) | 0.663 (0.127) | 0.581 (0.098) |
|                      | others     | 0.49 (0.69)  | 1.18 (1.03)  | 3016.03 (4.84)| 0.999 (0.000) | 0.997 (0.001) | 0.998 (0.001) |
| 20,000               | QC         | 9.39 (2.69)  | 2.85 (1.89)  | 3.92 (2.12)  | 0.585 (0.128) | 0.739 (0.118) | 0.643 (0.101) |
|                      | AC         | 2.74 (1.68)  | 8.80 (2.68)  | 4.78 (2.53)  | 0.548 (0.133) | 0.688 (0.136) | 0.597 (0.107) |
|                      | others     | 0.64 (0.83)  | 1.26 (1.08)  | 4015.62 (5.11)| 1.000 (0.000) | 0.998 (0.001) | 0.999 (0.000) |
| 25,000               | QC         | 9.29 (2.68)  | 2.64 (1.57)  | 3.83 (1.77)  | 0.593 (0.122) | 0.728 (0.138) | 0.644 (0.107) |
|                      | AC         | 2.84 (1.77)  | 7.70 (2.27)  | 5.75 (2.51)  | 0.479 (0.129) | 0.677 (0.133) | 0.550 (0.116) |
|                      | others     | 0.71 (0.85)  | 1.16 (1.08)  | 5016.08 (4.96)| 1.000 (0.000) | 0.998 (0.001) | 0.999 (0.000) |
| 30,000               | QC         | 9.45 (2.77)  | 2.65 (1.61)  | 4.20 (1.83)  | 0.583 (0.123) | 0.750 (0.117) | 0.648 (0.102) |
|                      | AC         | 2.32 (1.39)  | 7.37 (2.30)  | 5.66 (2.47)  | 0.483 (0.122) | 0.642 (0.145) | 0.542 (0.113) |
|                      | others     | 0.83 (0.93)  | 1.45 (1.19)  | 6016.07 (4.79)| 1.000 (0.000) | 0.998 (0.000) | 0.999 (0.000) |
Phase diagram

Figure S1: Predicted and experimentally determined phase diagrams for the 30 Al-TM-TM alloy systems. For each system, the left panel shows the predicted diagram made by the random forest classifier, and the right panel shows the actual phase diagram extracted from the literature. Three different colors are used to distinguish the phase region of quasicrystals, approximants, and others.
Machine learning learned rules

Figure S2: The formation rules of QC/AC discovered by the machine learning algorithm are overwritten on the predicted phase diagrams of the 30 Al-TM-TM systems. A color-coded line represents a condition imposed on the weighted average of van der Waals radius, electronegativity, first ionisation energy, the number of filled p valence orbitals, or the energy per atom at T=0K ground state.
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