**General Methods**

All manipulations were carried out under an inert atmosphere of dried nitrogen using standard Schlenk techniques. The Ag salts \( \text{Ag}^1 \text{Al} \{ \text{OC(CF}_3)_3 \}_4 \}_2 \) and \( \text{Ag}^1 \text{FAi}[\text{OC(C}_6\text{F}_5)(\text{C}_8\text{F}_{10})]_3 \}_2 \) were prepared according to literature procedures\(^{[1,2]}\). Solvents were freshly distilled under argon from \( \text{CaH}_2 \) (CH\(_2\)Cl\(_2\)) and from Na, K or Na/K alloy (toluene, n-pentane). IR spectra were recorded as KBr discs on Varian FTS-800 spectrometer. \(^1\)H, \(^{13}\)C, and \(^{31}\)P spectra were recorded on a Bruker Avance 300 spectrometer. \(^1\)H and \(^{13}\)C NMR chemical shifts were reported in parts per million (ppm) relative to Me\(_4\)Si as external standard. \(^{31}\)P NMR chemical shifts were expressed in ppm relative to external 85% H\(_3\)PO\(_4\) and were decoupled from the proton. Mass spectra were recorded by the MS department of the University of Regensburg on a ThermoQuest Finnigan TSQ 7000 mass spectrometer (ESI) or a Finnigan MAT 95 mass spectrometer (EI, FD).

**Synthesis of Ag\(_2\)(μ-\(\eta^1\)-1)\(\eta^2\)-1)\(\eta^2\)-1)[TEF]\(_2\) (2):**

\[
\begin{align*}
[\text{CpMo}(\text{CO})_4(\text{η}^2-\text{P}_2)] (1) & (50\text{mg, 0.1 mmol, 2 eq.}) \text{ was dissolved in 10 ml of CH}_2\text{Cl}_2. \\
\text{Ag(CH}_2\text{Cl}_2)[\text{TEF}] (A) & (58\text{ mg, 0.05 mmol, 1 eq.}) \text{ was dissolved in 5 mL of CH}_2\text{Cl}_2 \text{ and slowly added to the stirred solution of 1. The red solution is stirred for 2 h at room temperature, the solution was filtrated and carefully layered with 10 ml of n-pentane. In two days orange crystals of 7 are obtained, collected, washed with n-pentane (5 ml × 2) and dried in vacuum. Yield 90 mg (87%).} \\
\text{1H NMR (300 MHz, CD}_3\text{CN):} & \delta = 5.40 \text{ ppm (s, H}_{\text{Cp}}). \\
\text{13C}\{\text{1H} \} \text{ NMR (75.47 MHz, CD}_3\text{CN):} & \delta = 86.9 \text{ (s, C}_{\text{Cp}}), 120.8 \text{ (q, } J_{\text{FC}} = 289 \text{ Hz; C}_{\text{CF}_3}), 222.6 \text{ ppm (s, C}_\text{CO}). \\
\text{31P}\{\text{1H} \} \text{ NMR (121.49 MHz, CD}_3\text{CN):} & \delta = -86.1 \text{ ppm. ESI-MS (CH}_3\text{CN): m/z (\%) = 1100.6 (100) [Ag(C}_3\text{Mo}_2(\text{CO})_4(P)_2]_2^+. \text{ Elemental analysis, calcd. (\%) for C}_{88}\text{H}_{161}\text{Ag}_{2}\text{Al}_{7}\text{Mo}_{8}\text{N}_{2}\text{O}_{24}P_{8}: C 25.57, H 0.98, found: C 25.41, H 1.18. IR (KBr): } v/cm^-1 = 1989 \text{ (vs, 1958 (vs, 1426 (w, 1356 (s, 1304 (vs, 1277 (vs, 1242 (vs, 1219 (vs, 1171 (m, 1066 (w, 1012 (m, 973 (vs, 829 (s, 754 (w, 727 (vs, 560 (m), 537 (m), 490 (m), 443 (s).} \\
\text{Note: In the } ^{13}\text{C}\{\text{1H} \} \text{ NMR spectra of compounds 2 and 3 not all the signals from the [TEF] anions are detected.}
\end{align*}
\]
Synthesis of \([\text{Ag}_2(\mu-\eta^1:\eta^1-1)_3][\text{TEF}]_{2n}(3)\):

\[\text{CpMo}_2(\text{CO})_4(\eta^2-P_2)\] (1) (75 mg, 0.15 mmol, 1.5 eq.) was dissolved in 10 mL of CH$_2$Cl$_2$ and slowly added to Ag(CH$_2$Cl$_2$)[TEF] (A) (116 mg, 0.1 mmol, 1 eq.) in 5 mL of CH$_2$Cl$_2$. After 12 h stirring at room temperature the clear red solution was filtered and layered with the eightfold amount of toluene. After one day at room temperature compound 3 already crystallizes as red plates at the solvent mixing zone. Storage at +4 °C for three weeks resulted in further crystalline product. The supernatant solution was decanted off, the crystals were washed two times with toluene and dried in vacuum. Yield 127 mg (70%).

\[\text{H NMR} (300 \text{ MHz, CD}_3\text{CN}): \delta = 5.42 \text{ ppm} (s, \text{H}_{\text{Cp}}). \text{C}^{13}(^1\text{H}) \text{ NMR} (75.47 \text{ MHz, CD}_3\text{CN}): \delta = 87.0 \text{ (s, C}_{\text{Cp}}), 120.9 \text{ (q, J}_{\text{FC}} = 293 \text{ Hz}; C_{\text{CF}_3}), 222.2 \text{ ppm (s, C}_{\text{CO}}). \text{P}^{31}(^1\text{H}) \text{ NMR} (121.49 \text{ MHz, CD}_3\text{CN}): \delta = -93.0 \text{ ppm. ESI-MS (CH}_2\text{Cl}_2): m/z (\%) = 1100.7 (100). [\text{Ag}(\text{Cp}_2\text{Mo}_2(\text{CO})_4\text{P}_2)]^+, \text{Elemental analysis, calcd.} (\%) \text{ for } [\text{Ag}_2(\text{Cp}_2\text{Mo}_2(\text{CO})_4\text{P}_2)_3][\text{TEF}]_{2n}(\text{C}_7\text{H}_8): \text{C}, 26.00; \text{H}, 1.02; \text{found: C}, 26.04; \text{H}, 1.09. \text{IR (KBr)}: \tilde{\nu}/\text{cm}^{-1}: 3129 (w), 1993 (vs), 1968 (vs), 1942 (s), 1426 (w), 1353 (m), 1303 (vs), 1276 (vs), 1242 (vs), 1219 (vs), 1172 (m), 1066 (vw), 974 (vs), 831 (m), 728 (s), 559 (vw), 510 (vw), 453 (w).

Synthesis of \([\text{Ag}_2(\mu-\eta^1:\eta^1-1)_2(\eta^1-\text{CH}_2\text{Cl}_2)_{2}(\eta^2-\text{C}_7\text{H}_8)_2])[\text{FAI}]_{2}(4)\):

\[\text{CpMo}_2(\text{CO})_4(\eta^2-P_2)\] (1) (25 mg, 0.05 mmol, 1 eq.) was dissolved in 10 mL of CH$_2$Cl$_2$. Ag(CH$_2$Cl$_2$)[FAI] (B) (79 mg, 0.05 mmol, 1 eq.) was dissolved in 5 mL of CH$_2$Cl$_2$ and slowly added to the stirred solution of 1. The red solution is stirred for 2 h at room temperature, filtered and carefully layered with the fourfold amount of toluene. Storage at +4 °C affords orange crystals of 4 at the solvent mixing zone after several days. The crystals were washed with toluene and dried in vacuum. Yield 39 mg (39%). \text{H NMR} (300 MHz, CD$_3$CN): \delta = 5.42 ppm (s, H$_{\text{Cp}}$). \text{C}^{13}(^1\text{H}) \text{ NMR} (75.47 MHz, CD$_3$CN): \delta = 86.9 (s, C$_{\text{Cp}}$), 222.8 ppm (s, C$_{\text{CO}}$). \text{P}^{31}(^1\text{H}) \text{ NMR} (121.49 MHz, CD$_3$CN): \delta = -82.7 ppm. ESI-MS (CH$_2$Cl$_2$): m/z (\%) = 1100.6 (100). [Ag(Cp$_2$Mo$_2$(CO)$_4$P$_2$)$_3$]$^+$, \text{Elemental analysis, calcd.} (\%) \text{ for } [\text{Ag}(\text{Cp}_2\text{Mo}_2(\text{CO})_4\text{P}_2)_3(\text{C}_7\text{H}_8)$_3$][\text{FAI}]: \text{C}, 30.88; \text{H}, 0.76; \text{found: C}, 30.88; \text{H}, 1.15. \text{IR (KBr)}: \tilde{\nu}/\text{cm}^{-1}: 3122 (w), 2007 (vs), 1968 (vs), 1652 (w), 1533 (m), 1486 (vs), 1422 (w), 1323 (w), 1308 (w), 1267 (m), 1242 (m), 1204 (vs), 1187 (s), 1154 (m), 1135 (w), 1104 (m), 1033...
(w), 1019 (s), 1006 (m), 955 (vs), 911 (w), 838 (w), 770 (w), 729 (w), 635 (vw), 625 (vw), 457 (vw).

**Note:** In the $^{13}$C($^1$H) NMR spectra of compounds 4-6 the signals from the [FAI] anions are excluded due to two reasons: i) Only few signals are observed, ii) a very complicated set of couplings between C and F atoms is expected and its analysis is not the subject of this communication.

**Synthesis of $[Ag_2(\mu-n^1-1)_2(n^2-1)_2][FAI]_2$(5):**

$[\text{CpMo}_2(\text{CO})_4(n^2-P_2)]$ (1) (50mg, 0.1 mmol, 1 eq) was dissolved in 10 mL of CH$_2$Cl$_2$. Ag(CH$_2$Cl)$_2$[FAI] (B) (79 mg, 0.05 mmol, 1 eq.) was dissolved in 5 mL of CH$_2$Cl$_2$ and slowly added to the stirred solution of 1. The red solution is stirred for 2 h at room temperature, filtrated and carefully layered with the fourfold amount of toluene. Storage at +4 °C affords compound 5 as clear red prisms during one week. The crystals were washed with n-hexane and dried in vacuum. Yield 95 mg (76%). $^1$H NMR (300 MHz, CD$_3$CN): $\delta = 5.40$ ppm (s, H$_{Cp}$). $^{13}$C($^1$H) NMR (75.47 MHz, CD$_3$CN): $\delta = 87.1$ (s, C$_{Cp}$), 222.3 ppm (s, C$_{CO}$). $^{31}$P($^1$H) NMR (121.49 MHz, CD$_3$CN): $\delta = -92.6$ ppm. ESI-MS (CH$_2$Cl$_2$): m/z (%) = 1100.7 (100) $[Ag[Cp_2Mo_2(\text{CO})_4P_2]_2]^+$, Elemental analysis, calcd. (%) for $[Ag[Cp_2Mo_2(\text{CO})_4P_2]_2][\text{FAI}]$: C, 30.98; H, 0.81; found: C, 30.75; H, 1.02. IR (KBr): $\tilde{v}$/cm$^{-1}$: 3132 (w), 2008 (vs), 1981 (vs), 1940 (vs), 1652 (w), 1532 (m), 1485 (vs), 1424 (w), 1327 (w), 1267 (m), 1243 (m), 1204 (vs), 1186 (s), 1154 (m), 1136 (w), 1105 (m), 1034 (w), 1018 (s), 956 (vs), 910 (w), 824 (w), 767 (w), 752 (vw), 729 (w), 635 (vw), 559 (vw), 455 (vw).

**Synthesis of $[Ag_2(\mu-n^1-1)_4][FAI]_2n$(6):**

$[\text{CpMo}_2(\text{CO})_4(n^2-P_2)]$ (1) (50mg, 0.1 mmol, 1 eq.) was dissolved in 10 mL of CH$_2$Cl$_2$ and slowly added to a stirred solution of Ag(CH$_2$Cl)$_2$[FAI] (B) (79 mg, 0.05 mmol, 1 eq.) in 5 ml CH$_2$Cl$_2$. After 12 h stirring at room temperature the clear red solution was filtered and carefully layered with 20 ml of n-pentane. Within three days orange crystals of 6 are obtained, collected, washed with n-pentane (5 ml x 2) and dried in vacuum. Yield 112 mg (89%). $^1$H NMR (300 MHz, CD$_3$CN): $\delta = 5.38$ ppm (s, H$_{Cp}$). $^{13}$C($^1$H) NMR (75.47 MHz, CD$_3$CN): $\delta = 86.7$ (s, C$_{Cp}$), 223.5 ppm (s, C$_{CO}$). $^{31}$P($^1$H) NMR (121.49 MHz, CD$_3$CN): $\delta = -74.6$ ppm. ESI-MS (CH$_2$Cl$_2$): m/z (%)
= 1100.9 (100) [Ag(Cp₂Mo₂(CO)₄P₂)₂]⁺. Elemental analysis, calcd. (%) for [Ag(Cp₂Mo₂(CO)₄P₂)₂][FAI] C, 30.98; H, 0.81; found: C, 31.24; H, 0.92. IR (KBr): ν/cm⁻¹: 3128 (w), 2009 (vs), 1977 (vs), 1942 (vs), 1650 (w), 1532 (m), 1486 (vs), 1424 (w), 1325 (w), 1308 (w), 1266 (m), 1247 (m), 1200 (vs), 1187 (s), 1150 (m), 1136 (w), 1104 (m), 1032 (w), 1020 (s), 1001 (s), 910 (w), 828 (w), 771 (w), 730 (w), 635 (w), 593 (vw), 548 (vw), 456 (vw).

Crystallographic data

Crystals suitable for single crystal X-ray diffraction analysis were obtained for 2-6 as described above. The crystals were taken from a Schlenk flask under a stream of argon and immediately covered with perfluorinated Fomblin® mineral oil to prevent both decomposition and a loss of solvent. The quickly chosen single crystals covered by a drop of the oil were taken to the pre-centered goniometer head with CryoMount® and directly placed to the diffractometer into a stream of cold nitrogen.

Intensities were acquired using narrow ω-scans either an Oxford Diffraction Gemini R Ultra diffractometer equipped with micro-focus CuKα X-ray source and an AtlasS² CCD detector, or an Agilent Technologies SuperNova diffractometer equipped with a micro-focus CuKα source with an AtlasS² CCD detector (Tables S1-S2). All measurements were performed at 123 K, excepting 3, for which T = 100 K was used at the attempt to freeze out expected disorder. The data were processed with the CryAlis software package. An analytical absorption correction from crystal faces was applied for 2 and 4. A semi-empirical numerical absorption correction based on Gaussian integration over a multifaceted crystal model was applied for 3, 5 and 6.

Structure solution was performed by charge-flipping method with the program SUPERFLIP (4) or by direct methods with the program SHELXS or SHELXT (2, 3, 5 and 6). All structures were refined by full-matrix least-squares based on \( F^2 \) with different versions of program SHELXL running under Olex shell. All non-hydrogen atoms fully occupying their positions were refined with anisotropic displacement parameters. Hydrogen atoms were placed in idealized positions and refined isotropically according to the riding model. Figures were created with PLATON and Olex.

The crystal of 2 proved to be a two-component pseudo merohedral twin (twin law (-1 0 0, 0 1 0, 0 0 1)) of the monoclinic structures with a monoclinic angle of 90°. The twin batches were refined to 0.5/0.5. The only alternative orthorhombic space group \( Pmc2_1 \), was tried, but no satisfactory structure solution was found. The only systematic extinctions for \( 2_1(z) \) axis appear to be not independent, because they are the result from those for the \( c(y) \) glide plane. Therefore, twinned model seems to be correct.

In 3, the TEF anion as well as one Mo₃P₂ complex are severely disordered even at 100K. The Mo₃P₂ complex is disordered over 3 very close positions with occupancies refined as 0.53, 0.235 \( \times \) 2. The solvent toluene and CH₂Cl₂ molecules partly occupy their positions. During the refinement process the geometry of these groups had to be restrained.

In the crystal structure of 5 the positions of disordered CH₂Cl₂ and toluene molecules mostly overlap. The molecular occupancy factors vary from 0.13 to 0.67. Two least occupied positions of toluene (0.13 and 0.16) molecules were refined with geometry restraints. The rest solvate molecules were refined freely after fixing their occupancy factors.

CCDC reference numbers 1551486 (2), 1551487 (3), 1551488 (4), 1551489 (5), and 1551490 (6) contain the supplementary crystallographic data, which can be obtained free of charge at
Table S1: Structure determination summary of complexes 2, 3 and 4

|                  | 2                      | 3                      | 4                      |
|------------------|------------------------|------------------------|------------------------|
| Crystal data     |                        |                        |                        |
| CCDC             | 1551486                | 1551487                | 1551488                |
| Chemical formula | C₁₈₀₈₉.₅₀H₄₃.₄₂Ag₂Cl₂F₂Mo₁₈₂P₈ | C₁₈₁H₁₇₂₀⁺Ag₂Al₂Cl₉₂F₂₂O₅₂P₆ | C₈₅H₉₂AgCl₃F₁₈₂Mo₃₂O₆P₃ |
| Mᵣ              | 4261.57                | 3745.82                | 2162.31                |
| Crystal system, space group | Monoclinic, P2₁/c | Monoclinic, P2₁/c | Monoclinic, P2₁/c |
| Temperature (K)  | 123                    | 100                    | 123                    |
| α, b, c (Å)      | 10.2377 (1), 21.3490 (2), 59.4112 (5) | 19.44134 (16), 17.47231 (13), 34.8842 (2) | 12.8202 (2), 19.0679 (4), 27.8091 (4) |
| α, β, γ (°)      | 90, 90.022 (1), 90 | 101.9624 (7) | 95.734 (1) |
| V (Å³)           | 12985.2 (2)            | 11592.33 (15)          | 6764.0 (2)             |
| Z                | 4                      | 4                      | 4                      |
| F(000)           | 8188                   | 7203                   | 4184                   |
| Dₐ (Mg m⁻³)      | 2.180                  | 2.146                  | 2.123                  |
| Radiation type   | Cu Kα                  | Cu Kα                  | Cu Kα                  |
| µ (mm⁻¹)         | 11.66                  | 10.55                  | 8.29                   |
| Crystal colour, habit | Orange plate | light red prism | Light orange |
| Crystal size (mm) | 0.23 × 0.16 × 0.07    | 0.22 × 0.15 × 0.14    | 0.15 × 0.15 × 0.04    |
| Data collection  |                        |                        |                        |
| Diffractometer   | Xcalibur, Atlas, Gemini ultra | Xcalibur, Atlas, Gemini ultra | SuperNova, Single source at offset, AtlasS2 |
| Absorption correction | Analytical          | Gaussian               | Analytical             |
| Tₘin, Tₚax      | 0.121, 0.433           | 0.229, 0.406           | 0.483, 0.779           |
| No. of measured, independent and observed [I > 2σ(I)] reflections | 93086, 23166, 21809 | 26464, 10155, 8861 | 23992, 13025, 10379 |
| Rint            | 0.055                  | 0.020                  | 0.042                  |
| (sin θ/λ)ₚax (Å⁻¹) | 0.602                | 0.597                  | 0.621                  |
| Range of h, k, l | h = -11→12, k = -25→25, l = -69→70 | h = -21→22, k = -18→20, l = -41→32 | h = -15→12, k = -23→23, l = -25→34 |
| Refinement       |                        |                        |                        |
| R(F²) > 2σ(F²), wR(F²), S | 0.044, 0.119, 1.09 | 0.058, 0.171, 1.06 | 0.051, 0.143, 1.02 |
| No. of reflections | 23166                | 10155                  | 13025                  |
| No. of parameters | 1925                | 1180                   | 1073                   |
| No. of restraints | 5                   | 312                    | 0                      |
| H-atom treatment | H-atom parameters constrained | H-atom parameters constrained | H-atom parameters constrained |
| Δ(Δmax, Δmin) (e Å⁻³) | 1.98, -1.20          | 1.60, -1.76           | 2.74, -1.34            |

Computer programs: CrysAlis PRO, Agilent Technologies, Version 1.171.37.31d (release 11-02-2014), SHELXS, SHELXL2013 (Sheldrick, 2013), SHELXT (Sheldrick, 2015), SHELXL (Sheldrick, 2008), Olex2 (Dolomanov et al., 2009), (Palatinus & Chapuis, 2007; Palatinus & van der Lee, 2008; Palatinus et al., 2012).
### Table S2: Structure determination summary of complexes 5 and 6

|                  | 5                                            | 6                                            |
|------------------|----------------------------------------------|----------------------------------------------|
| **Crystal data** |                                              |                                              |
| CCDC             | 1551489                                      | 1551490                                      |
| Chemical formula | C_{165.86}H_{83.44}Ag_{2}Cl_{0.40}Mo_{8}O_{22}F_{46}P_{8} | C_{28}H_{20}AgMo_{4}O_{8}·AlC_{36}F_{46}O_{3}·CH_{2}Cl_{2} |
| \(M_r\)         | 5475.24                                      | 2566.21                                      |
| Crystal system,  | Monoclinic, \(C2/c\)                        | Monoclinic, \(C2/c\)                        |
| space group      |                                              |                                              |
| Temperature (K)  | 123                                          | 123                                          |
| \(a, b, c\) (Å)  | 29.9499 (4), 21.10827 (19), 33.3307 (4)      | 41.0189 (7), 11.2730 (1), 39.8120 (7)        |
| \(\alpha, \beta, \gamma\) (°) | 113.6385 (15) | 122.547 (2) |
| \(V\) (Å³)      | 19303.3 (4)                                  | 15518.1 (5)                                  |
| \(Z\)           | 4                                            | 8                                            |
| \(F(000)\)      | 10662                                        | 9888                                         |
| \(D_r\) (Mg m\(^{-3}\)) | 1.884                                       | 2.197                                        |
| Radiation type   | Cu \(\text{K}\alpha\)                      | Cu \(\text{K}\alpha\)                      |
| \(\mu\) (mm\(^{-1}\)) | 7.85                                        | 10.26                                        |
| Crystal colour,  | Light red prism                             | Orange prism                                |
| habit            |                                              |                                              |
| Crystal size (mm) | 0.12 × 0.10 × 0.06                          | 0.34 × 0.17 × 0.05                          |
| **Data collection** |                                              |                                              |
| Diffractometer   | SuperNova, Single source at offset, Atlas    | SuperNova, Titan\(^{22}\)                    |
| Absorption       | Gaussian                                     | Gaussian                                     |
| correction       |                                              |                                              |
| \(T_{\min}, T_{\max}\) | 0.737, 0.842                                | 0.133, 0.662                                |
| No. of measured,  | 35747, 18313, 13475                          | 27184, 15012, 14406                         |
| independent and  |                                              |                                              |
| observed \([I > 2\sigma(I)]\) reflections |                                              |                                              |
| \(R_{int}\)     | 0.027                                        | 0.034                                        |
| \(\sin \theta_{\text{max}}\) (Å\(^{-1}\)) | 0.618                                       | 0.624                                        |
| Range of \(h, k, l\) | \(h = -36 \rightarrow 35, k = -16 \rightarrow 25, l = -39 \rightarrow 41\) | \(h = -50 \rightarrow 50, k = -13 \rightarrow 13, l = -41 \rightarrow 49\) |
| **Refinement**   |                                              |                                              |
| \(R(F^2 > 2\sigma(F^2)), wR(F^2), S\) | 0.040, 0.104, 0.93                         | 0.042, 0.119, 1.06                         |
| No. of reflections | 18313                                        | 15012                                        |
| No. of parameters | 1444                                         | 1220                                         |
| No. of restraints | 2                                            | 0                                            |
| H-atom treatment | H-atom parameters constrained                | H-atom parameters constrained                |
| \(\Delta_{\text{max}}, \Delta_{\text{min}}\) (Å\(^{-3}\)) | 1.24, -1.07                                 | 1.68, -1.81                                 |

Computer programs: *CrysAlis PRO* 1.171.38.41h (Rigaku OD, 2015), *SHELXT* (Sheldrick, 2015), *SHELXL* (Sheldrick, 2015), Olex2 (Dolomanov et al., 2009).
Figure S1: Molecular structure of a dication (top) and two independent TEF anions (bottom) in 2 in the solid state, showing the atom-labelling scheme. Displacement ellipsoids are drawn at the 50% probability level (hydrogen atoms omitted for clarity, disordered groups are shown in dashed lines).
### Table S3: Selected geometric parameters (Å, °) for 2

| Bond          | Distance  | Bond          | Distance  |
|---------------|-----------|---------------|-----------|
| Ag1—P12       | 2.4634 (16)| Mo22—C222     | 2.375 (7) |
| Ag1—P22       | 2.4916 (16)| Mo22—P21      | 2.4706 (18)|
| Ag1—P31       | 2.5709 (17)| Mo22—P22      | 2.526 (2)  |
| Ag1—P32       | 2.6679 (17)| Mo31—C31A     | 2.013 (7)  |
| Ag2—P21       | 2.4841 (17)| Mo31—C31B     | 2.015 (9)  |
| Ag2—P11       | 2.5200 (16)| Mo31—C314     | 2.272 (7)  |
| Ag2—P41       | 2.5990 (17)| Mo31—C315     | 2.311 (8)  |
| Ag2—P42       | 2.6885 (17)| Mo31—C313     | 2.340 (6)  |
| Mo31—C11B     | 1.992 (8)  | Mo31—C311     | 2.362 (7)  |
| Mo31—C11A     | 2.010 (8)  | Mo31—C312     | 2.368 (7)  |
| Mo31—C114     | 2.280 (8)  | Mo31—P31      | 2.4379 (16)|
| Mo31—C113     | 2.297 (8)  | Mo31—P32      | 2.5234 (17)|
| Mo31—C115     | 2.338 (9)  | Mo31—Mo32     | 3.0266 (7) |
| Mo31—C112     | 2.359 (8)  | Mo32—C32B     | 1.982 (7)  |
| Mo31—C111     | 2.370 (8)  | Mo32—C32A     | 2.008 (9)  |
| Mo31—P12      | 2.4363 (18)| Mo32—C323     | 2.296 (7)  |
| Mo31—P11      | 2.538 (2)  | Mo32—C322     | 2.317 (8)  |
| Mo31—Mo12     | 3.0236 (8) | Mo32—C324     | 2.329 (6)  |
| Mo12—C12B     | 2.013 (8)  | Mo32—C321     | 2.354 (7)  |
| Mo12—C12A     | 2.015 (10) | Mo32—C325     | 2.369 (6)  |
| Mo12—C125     | 2.293 (9)  | Mo32—P32      | 2.4575 (17)|
| Mo12—C124     | 2.296 (9)  | Mo32—P31      | 2.5261 (16)|
| Mo12—C123     | 2.333 (8)  | Mo41—C41A     | 1.996 (7)  |
| Mo12—C121     | 2.363 (8)  | Mo41—C41B     | 2.035 (8)  |
| Mo12—C122     | 2.404 (9)  | Mo41—C415     | 2.290 (6)  |
| Mo12—P11      | 2.4613 (16)| Mo41—C411     | 2.320 (7)  |
| Mo12—P12      | 2.5087 (16)| Mo41—C414     | 2.327 (6)  |
| C241—Mo21     | 2.33 (2)   | Mo41—C412     | 2.360 (6)  |
| C242—Mo21     | 2.326 (18) | Mo41—C413     | 2.368 (6)  |
| C243—Mo21     | 2.272 (17) | Mo41—P41      | 2.4430 (16)|
| C244—Mo21     | 2.26 (2)   | Mo41—P42      | 2.5345 (17)|
| C245—Mo21     | 2.32 (3)   | Mo41—Mo42     | 3.0252 (7) |
| C21B—Mo21     | 2.04 (2)   | Mo42—C42A     | 1.989 (8)  |
| C231—Mo21     | 2.364 (11) | Mo42—C42B     | 2.015 (9)  |
| C232—Mo21     | 2.410 (16) | Mo42—C423     | 2.284 (7)  |
| C233—Mo21     | 2.326 (12) | Mo42—C422     | 2.309 (7)  |
| C234—Mo21     | 2.316 (11) | Mo42—C424     | 2.334 (7)  |
| C235—Mo21     | 2.305 (11) | Mo42—C421     | 2.340 (8)  |
| C21A—Mo21     | 2.038 (14) | Mo42—C425     | 2.359 (8)  |
| Mo21—C21C     | 2.009 (7)  | Mo42—P42      | 2.4483 (17)|
| Mo22—C22A     | 1.979 (7)  | Mo42—P41      | 2.5251 (17)|
| Mo22—C22B     | 2.025 (7)  | P11—P12       | 2.087 (2)  |
| Mo22—C225     | 2.284 (7)  | P21—P22       | 2.088 (2)  |
| Mo22—C224     | 2.290 (7)  | P31—P32       | 2.152 (3)  |
| Mo22—C221     | 2.348 (7)  | P41—P42       | 2.149 (2)  |
| Mo22—C223     | 2.362 (7)  |               |            |
Figure S2: Independent part of a cation in 3 in the solid state, showing the atom-labelling scheme. Displacement ellipsoids are drawn at the 50% probability level. (hydrogen atoms omitted for clarity, disordered fragments are shown in ball-and-stick model).
Table S4: Selected geometric parameters (Å, º) for 3

| Bond | Å    | Bond | Å    |
|------|------|------|------|
| Ag1—P1 | 2.4464 (14) | P3—Mo4 | 2.502 (6) |
| Ag1—P2 | 2.4642 (15) | P3—Mo3 | 2.310 (16) |
| Ag1—P3 | 2.4425 (16) | P3—Mo3 | 2.420 (17) |
| Mo1—Mo2 | 3.0615 (8) | C20—Mo4 | 1.991 (12) |
| Mo1—P1 | 2.5199 (16) | C21—Mo5 | 2.02 (6) |
| Mo1—P2 | 2.4461 (16) | C121—Mo4 | 2.014 (17) |
| Mo1—Mo2 | 2.303 (8) | Mo5—P3 | 2.683 (17) |
| Mo1—C2AA | 2.321 (8) | Mo5—C225 | 2.27 (3) |
| Mo1—C3AA | 2.334 (8) | Mo5—C226 | 2.31 (4) |
| Mo1—C4AA | 2.367 (8) | Mo5—C227 | 2.34 (3) |
| Mo1—C5 | 2.327 (7) | Mo5—C228 | 2.26 (3) |
| Mo1—C6AA | 2.004 (9) | Mo5—C229 | 2.25 (3) |
| Mo1—C7 | 1.993 (9) | Mo5—C220 | 2.014 (19) |
| Mo2—P1 | 2.4279 (16) | Mo4—P3 | 2.502 (6) |
| Mo2—P2 | 2.5212 (17) | Mo4—Mo4 | 3.034 (9) |
| Mo2—C8 | 2.352 (7) | Mo4—C15 | 2.300 (15) |
| Mo2—C9 | 2.312 (7) | Mo4—C16 | 2.298 (17) |
| Mo2—C1AA | 2.306 (9) | Mo4—C17 | 2.339 (16) |
| Mo2—C11 | 2.325 (9) | Mo4—C18 | 2.379 (16) |
| Mo2—C12 | 2.345 (8) | Mo4—C19 | 2.355 (15) |
| Mo2—C13 | 2.005 (8) | Mo3—P3 | 2.420 (17) |
| Mo2—C14 | 1.991 (8) | Mo3—Mo3 | 2.16 (3) |
| P1—Mo1 | 2.5198 (16) | Mo3—C115 | 2.32 (3) |
| P1—Mo2 | 2.4279 (16) | Mo3—C116 | 2.38 (3) |
| P1—P2 | 2.097 (2) | Mo3—C117 | 2.35 (3) |
| P2—P1 | 2.097 (2) | Mo3—C118 | 2.33 (3) |
| P3—P3 | 2.082 (3) | Mo3—C119 | 2.33 (3) |
| P3—Mo5 | 2.683 (17) | Mo3—C120 | 2.008 (18) |
| P3—Mo5 | 2.632 (16) | Mo3—C221 | 2.02 (5) |
| P3—Mo4 | 2.410 (6) | |

Symmetry code(s): (i) -x+1, -y+2, -z+1; (ii) -x+1/2, y, -z+1.

Table S5: Selected geometric parameters (Å, º) for 4

| Bond | Å    | Bond | Å    |
|------|------|------|------|
| Ag1—Ag1 | 3.0532 (10) | Mo1—C7 | 2.352 (7) |
| Ag1—P1 | 2.4523 (14) | Mo2—P1 | 2.5028 (15) |
| Ag1—P2 | 2.4550 (13) | Mo2—P2 | 2.4479 (14) |
| Mo1—Mo2 | 3.0549 (6) | Mo2—C8 | 1.988 (7) |
| Mo1—P1 | 2.4438 (14) | Mo2—C9 | 1.994 (6) |
| Mo1—P2 | 2.5020 (12) | Mo2—C10 | 2.339 (7) |
| Mo1—C1 | 1.985 (7) | Mo2—C11 | 2.342 (6) |
| Mo1—C2 | 2.025 (7) | Mo2—C12 | 2.333 (6) |
| Mo1—C3 | 2.338 (8) | Mo2—C13 | 2.299 (6) |
| Mo1—C4 | 2.313 (7) | Mo2—C14 | 2.304 (6) |
| Mo1—C5 | 2.278 (6) | P2—Ag1 | 2.4550 (13) |
| Mo1—C6 | 2.323 (6) | |

Symmetry code(s): (i) -x, -y, -z+1.
Figure S3: Molecular structure of a cation (top) and an anion (bottom) of 4 in the solid state, showing the atom-labelling scheme. The coordinated CH$_2$Cl$_2$ and toluene molecules are omitted for clarity. Displacement ellipsoids are drawn at the 50% probability level. H atoms are omitted for clarity. Symmetry code: (a) - x, -y, 1 - z.
Figure S4: Molecular structure of a cation (top) and an anion (bottom) of 5 in the solid state, showing the atom-labelling scheme. Displacement ellipsoids are drawn at the 50% probability level. H atoms are omitted for clarity.
Table S6: Selected geometric parameters (Å, °) for 5

| Bond                  | Length (Å) | Angle (°)  |
|-----------------------|------------|------------|
| Ag1—P4                | 2.4834 (11) | Mo4—C30    | 2.304 (5) |
| Ag1—P3                | 2.4871 (11) | Mo4—C33    | 2.319 (5) |
| Ag1—P2                | 2.5988 (11) | Mo4—C32    | 2.340 (5) |
| Ag1—P1                | 2.7122 (11) | Mo4—C31    | 2.351 (5) |
| Mo1—C2                | 1.979 (6)   | Mo4—P4     | 2.4533 (11) |
| Mo1—C1                | 2.001 (5)   | Mo4—P4¹    | 2.5215 (11) |
| Mo1—C7                | 2.301 (4)   | P1—P2      | 2.1395 (16) |
| Mo1—C3                | 2.311 (5)   | P3—P3¹     | 2.091 (2)  |
| Mo1—C6                | 2.333 (4)   | P3—Mo3     | 2.4406 (11) |
| Mo1—C                  | 2.360 (5)   | P3—Mo3³    | 2.5285 (12) |
| Mo1—C5                | 2.379 (5)   | P3—Mo3³    | 2.5285 (12) |
| Mo1—P4                | 2.4642 (11) | P4—P4¹     | 2.090 (2)  |
| Mo1—P2                | 2.5374 (11) | P4—Mo4¹    | 2.5216 (11) |
| Mo1—Mo2               | 3.0206 (5)  | Mo3—C15    | 1.990 (3)  |
| Mo2—C9                | 1.999 (5)   | Mo3—C16    | 2.000 (5)  |
| Mo2—C8                | 2.010 (4)   | Mo3—C17    | 2.291 (9)  |
| Mo2—C12               | 2.294 (5)   | Mo3—C18    | 2.314 (9)  |
| Mo2—C13               | 2.306 (5)   | Mo3—C25    | 2.318 (10) |
| Mo2—C11               | 2.331 (5)   | Mo3—C26    | 2.323 (9)  |
| Mo2—C14               | 2.358 (5)   | Mo3—C23    | 2.327 (11) |
| Mo2—C10               | 2.369 (5)   | Mo3—C22    | 2.328 (10) |
| Mo2—P2                | 2.4464 (11) | Mo3—C21    | 2.329 (11) |
| Mo2—P1                | 2.5351 (11) | Mo3—C24    | 2.344 (10) |
| Mo4—C27               | 2.003 (5)   | Mo3—C19    | 2.371 (10) |
| Mo4—C28               | 2.013 (5)   | Mo3—C20    | 2.393 (12) |
| Mo4—C29               | 2.291 (5)   |  |  |

Symmetry code(s): (i) -x+1, y, -z+3/2.
Figure S5: Independent part of a cationic chain (top) and molecular structure of an FAL anion (bottom) in 6 in the solid state (hydrogen atoms omitted for clarity).
### Table S7: Selected geometric parameters (Å, °) for 6

| Bond                  | Distance (Å) | Bond Angle (°) |
|-----------------------|--------------|----------------|
| Ag1—P2                | 2.5775 (8)   |                |
| Ag1—P2<sup>i</sup>   | 2.5775 (8)   |                |
| Ag1—P3                | 2.5932 (8)   | 1.988 (4)      |
| Ag1—P3<sup>i</sup>   | 2.5933 (8)   | 1.992 (4)      |
| Ag2—P4                | 2.6059 (8)   | 2.311 (4)      |
| Ag2—P4<sup>i</sup>   | 2.6059 (8)   | 2.296 (4)      |
| Ag2—P1<sup>ii</sup>  | 2.6086 (8)   | 2.317 (4)      |
| Ag2—P1<sup>iii</sup> | 2.6086 (8)   | 2.341 (4)      |
| Mo1—C1                | 1.992 (4)    | 3.0367 (7)     |
| Mo1—C2                | 1.997 (4)    | 2.4565 (8)     |
| Mo1—C4                | 2.295 (3)    | 2.5297 (9)     |
| Mo1—C3                | 2.311 (4)    | 3.0367 (7)     |
| Mo1—C5                | 2.314 (4)    | 1.996 (4)      |
| Mo1—C7                | 2.351 (4)    | 1.996 (4)      |
| Mo1—C6                | 2.359 (4)    | 2.290 (4)      |
| Mo1—P1                | 2.4589 (8)   | 2.299 (4)      |
| Mo1—P2                | 2.5274 (8)   | 2.327 (4)      |
| Mo1—Mo2               | 3.0206 (4)   | 2.349 (4)      |
| Mo2—C8                | 1.979 (4)    | 2.368 (4)      |
| Mo2—C9                | 2.008 (4)    | 2.4564 (8)     |
| Mo2—C10               | 2.291 (4)    | 2.5295 (8)     |
| Mo2—C14               | 2.313 (4)    | 2.1005 (12)    |
| Mo2—C11               | 2.326 (4)    | 2.6086 (8)     |
| Mo2—C13               | 2.351 (4)    | 2.1004 (12)    |
| Mo2—C12               | 2.369 (4)    |                |

P2—Ag1—P2<sup>i</sup> 94.77 (4)          P1—Mo2—Mo1 51.65 (2)
P2—Ag1—P3    119.88 (3)          P4—Mo3—Mo4 53.58 (2)
P2<sup>i</sup>—Ag1—P3    114.81 (3)          P3—Mo3—Mo4 51.391 (18)
P2—Ag1—P3<sup>i</sup> 114.81 (3)          P3—Mo4—Mo3 53.59 (2)
P2<sup>i</sup>—Ag1—P3<sup>i</sup> 119.88 (3)        P4—Mo4—Mo3 51.396 (19)
P3—Ag1—P3<sup>i</sup> 94.66 (4)          P2—P1—Mo1 66.79 (3)
P4<sup>i</sup>—Ag2—P4 92.33 (4)          P2—P1—Mo2 63.12 (3)
P4<sup>i</sup>—Ag2—P1<sup>ii</sup> 116.23 (3)        P2—P1—Ag2<sup>iv</sup> 133.49 (4)
P4<sup>ii</sup>—Ag2—P1<sup>ii</sup> 121.81 (3)        P1—P2—Mo2 67.08 (3)
P4<sup>iii</sup>—Ag2—P1<sup>iii</sup> 116.23 (3)        P1—P2—Mo1 63.40 (3)
P4<sup>ii</sup>—Ag2—P1<sup>iiii</sup> 91.22 (4)         P1—P2—Ag1 130.04 (4)
P4<sup>iiii</sup>—Ag2—P1<sup>iiii</sup> 91.22 (4)         P4—P3—Mo4 66.91 (3)
P1—Mo1—Mo2 53.89 (2)          P4—P3—Mo3 63.29 (3)
P2—Mo1—Mo2 51.55 (2)          P4—P3—Ag1 130.40 (4)
C8—Mo2—P1 70.99 (12)           P3—P4—Mo3 66.91 (3)
C9—Mo2—P1 79.72 (13)           P3—P4—Mo4 63.29 (3)
P2—Mo2—P1 49.80 (3)           P3—P4—Ag2 132.96 (4)
P2—Mo2—Mo1 53.79 (2)

Symmetry code(s): (i) -x+1, y, -z+3/2; (ii) x, y+1, z; (iii) -x+1, y+1, -z+3/2; (iv) x, y-1, z.
DFT calculations

The DFT calculations have been performed with the TURBOMOLE program package\(^{10}\) at the RI-B3LYP/def2-TZVP\(^{13}\) level of theory. The geometries were optimized in the gas phase using the Multipole Accelerated Resolution of Identity (MARI-J)\(^{14}\) approximation during the geometry optimization steps. The solvent effects were incorporated as single point calculations (without the RI approximation) on the gas phase optimized geometries via the Conductor-like Screening Model (COSMO)\(^{15}\) using the dielectric constant of CH\(_2\)Cl\(_2\) (\(\epsilon = 8.930\)). For the reaction energies the SCF energies, corrected for the “outlying charge” were used.

### Table S8. Total Energies (a.u.) calculated at the B3LYP/def2-TZVP (\(\epsilon = 8.930\)) level of theory.\(^a\)

|       | C          | D          | I          |
|-------|------------|------------|------------|
| Tot. Energy | -6784.9331038083 | -5125.4223682595 | -1659.5155691591 |
| Tot. Energy + OC corrections | -6784.930196620 | -5125.419613819 | -1659.5158776158 |

### Table S9. Cartesian coordinates of the gas-phase optimized geometry of \([\{\text{Cp(CO)}_2\text{Mo}\}_2\{\mu-\text{P}_2\}]_2\text{Ag}_2\)^{2+} at the RI-B3LYP/def2-TZVP level of theory. (E = -6931.65150897001 a.u.).

| Atom | x        | y        | z        |
|------|----------|----------|----------|
| Ag   | 0.2667583| 2.3084358| 0.9091437|
| Ag   | -0.2661552| -2.3077761| -0.9133967|
| Mo   | 2.8016500| -1.4843509| 2.5543113|
| Mo   | -0.0023169| -1.4159941| 3.9231411|
| Mo   | 0.0021376| 1.4167346| -3.9240237|
| Mo   | -2.8019964| 1.4841757| -2.555183|
| Mo   | -0.1721732| 5.5453487| 3.9710873|
| Mo   | 0.4179510| 6.7492946| 1.1557833|
| Mo   | 0.1706909| -5.5466110| -3.9711883|
| Mo   | -0.4175390| -6.7485643| -1.1545810|
| P    | 0.4620435| -1.7815307| 1.5175874|
| P    | 0.9864416| 0.1476421| 2.1369227|
| P    | -0.9863814| -0.1476618| -2.1384335|

\(\text{Cp(CO)}_2\text{Mo}\) = cyclopentadienyl(1,5-diisobutyloctane)mo-lylene
\(\mu-\text{P}_2\) = μ-1,5-bis(diphenylphosphino)pentane
| Element | X    | Y    | Z    |
|---------|------|------|------|
| P       | -0.4626543 | 1.7815802 | -1.5185236 |
| P       | 1.1794019 | 4.5145835 | 2.1675551 |
| P       | -0.8939150 | 4.7015951 | 1.6579333 |
| P       | -1.1820463 | -4.5158340 | -2.1684849 |
| P       | 0.8915752 | -4.6995927 | -1.6599974 |
| C       | 0.4261134 | 3.7679074 | -4.1713385 |
| H       | 0.3823311 | 4.4636298 | -3.3509803 |
| C       | -0.6317508 | 3.4496196 | -5.0649678 |
| H       | -1.6192691 | 3.8773346 | -5.0541988 |
| C       | -0.1407284 | 2.5163593 | -6.0116634 |
| H       | -0.6911888 | 2.1096711 | -6.8448467 |
| C       | 1.2280776 | 2.2595736 | -5.7162132 |
| H       | 1.8966939 | 1.6389267 | -6.2898884 |
| C       | 1.5733274 | 3.0404583 | -4.5718007 |
| H       | 2.5482236 | 3.1010606 | -4.1159121 |
| O       | 2.6325298 | -0.1329781 | -3.1842205 |
| C       | 1.6483159 | 0.3935743 | -3.4221501 |
| O       | 2.4634583 | -4.6012636 | 2.2980740 |
| O       | -2.6333893 | 0.1320112 | 3.1820358 |
| O       | -3.6204388 | 1.4219916 | 0.4733374 |
| O       | -1.5519108 | 2.8313087 | 4.7103070 |
| O       | 3.3129700 | 7.3731371 | 2.2203342 |
| O       | 1.5473116 | -2.8318030 | -4.7133578 |
| O       | -1.7088708 | -5.3797752 | 1.3446022 |
| O       | 3.6193144 | -1.4232386 | -0.4747753 |
| O       | 1.0130000 | 1.0695957 | 5.5606631 |
| O       | -1.0140470 | -1.0671631 | -5.5637646 |
| O       | -2.4647618 | 4.6011195 | -2.2985416 |
| O       | -3.0609937 | 6.7501802 | 3.658269 |
| O       | 1.7081766 | 5.3812417 | -1.3443957 |
| O       | 3.0610305 | -6.7476553 | -3.6564249 |
| O       | -3.3122339 | -7.3762336 | -2.2175702 |
| C       | 2.5462479 | -3.4593206 | 2.3786006 |
| C       | 3.2707299 | -1.4479588 | 0.613513 |
| C       | 0.6484923 | 0.1853607 | 4.9267235 |
| C       | -1.6489557 | -0.3938993 | 3.4204435 |
| C       | -0.6492370 | -0.1836022 | -4.9291074 |
C -2.5471695 3.4591768 -2.3793240
C -3.2715059 1.4470742 -0.6146639
C -2.0082759 6.3027569 3.720164
C -1.0538889 3.8146373 4.3984955
C 1.2542929 5.8526318 -0.3999874
C 2.2591536 7.1043514 1.8593627
C 2.0077505 -6.3015878 -3.7199671
C 1.0503658 -3.8153510 -4.4005075
C -2.2585492 -7.1060766 -1.8572177
C -1.2545662 -5.8514004 0.4005179
C 3.7382518 -0.9115568 4.7087983
H 3.1880961 -0.8917432 5.6334018
C 3.9477211 0.1921507 3.8381629
H 3.5675391 1.1899671 3.9797547
C 4.7819893 -0.2367422 2.7769092
H 5.1557701 0.3811650 1.9765943
C 5.0884496 -1.6158947 2.9827701
H 5.7469857 -2.2187767 2.3793046
C 4.4339891 -2.0260430 4.1783038
H 4.5012721 -3.0034820 4.6285032
C 0.4268998 5.87670855 4.1725650
H 0.3845461 -4.4633147 3.3525477
C 0.6324069 -3.4485182 5.0643696
H 1.6197117 -3.8766824 5.0524914
C 0.1431688 -2.5143532 6.0111059
H 0.6949510 -2.1073501 6.8432652
C 1.2260104 -2.2573028 5.7175204
H 1.8935697 -1.6360540 6.2917848
C 1.5732355 -3.0388982 4.5742149
H 2.5488306 -3.0993955 4.1197873
C -4.4347204 2.0265492 -4.1789071
H -4.5026786 3.0044299 -4.6280490
C -3.7382024 0.9131307 -4.7106033
H -3.1881483 0.8946722 -5.6352983
C -3.9469030 -0.1916521 -3.8411172
H -3.5660872 -1.1890764 -3.9837722
C -4.7814954 0.2355272 -2.7794323
H  -5.1548304  -0.3835002  -1.9797633
C   -5.0888939  1.6146781  -2.9838258
H  -5.7478653  2.2164689  -2.3793719
C   1.9009737  5.9539331  5.1169920
H   2.8635475  5.6815595  4.7172991
C   1.2683737  7.2178741  4.9694926
H   1.6710509  8.0693411  4.4484537
C    0.0420777  7.1799111  5.6772075
H  -0.6535980  7.9969547  5.7828763
C   -0.0879874  5.8962440  6.2745986
H  -0.8858214  5.5775484  6.9251936
C    1.0708422  5.1373093  5.9238119
H    1.2922839  4.1343061  6.2506375
C   -1.6758752  7.8452462  0.7185182
H  -2.6319316  7.3629100  0.8350645
C   -0.8881713  7.8569315  -0.4579699
H  -1.1471682  7.3896466  -1.3939808
C    0.2772459  8.6435809  -0.2020400
H    1.0497024  8.8902152  -0.9119016
C    0.1920167  9.1137650  1.1364915
H    0.9031474  9.7628569  1.6220664
C   -1.0119857  8.6277367  1.7023991
H   -1.3814386  8.8512618  2.6882681
C   -1.0720508  -5.1419064  -5.9248751
H  -1.2937708  -4.1394925  -6.2533163
C   -1.9021410  -5.9575945  -5.1170627
H  -2.8649518  -5.6849790  -4.7181043
C   -1.2690865  -7.2210706  -4.9674717
H  -1.6716168  -8.0718971  -4.4452832
C   -0.0426102  -7.1837417  -5.6748768
H    0.6533651  -8.0007109  -5.7791310
C    0.0871697  -5.9009453  -6.2741847
H    0.8850608  -5.5829388  -6.9250461
C    1.6774336  -7.8417926  -0.7157083
H    2.6329543  -7.3582751  -0.8317765
C    0.8889364  -7.8541227  0.4602498
H    1.1467444  -7.3862425  1.3962933
20

C -0.2753307 -8.6422858 0.2037696
H -1.0479461 -8.8896968 0.9131886
C -0.1886292 -9.1126873 -1.1346057
H -0.8986094 -9.7628135 -1.6204784
C 1.0151673 -8.6253651 -1.6998054
H 1.3855937 -8.8487486 -2.6853328

Table S10. Cartesian coordinates of the gas-phase optimized geometry of [(Cp(CO)$_2$Mo)$_2$(μ-P$_2$)]$_4$Ag$^+$ at the RI-B3LYP/def2-TZVP level of theory. (E = -6784.871993216 a.u.).

| Atom | x    | y    | z    |
|------|------|------|------|
| Mo   | 2.5989743 | 2.5496424 | -3.3858428 |
| Mo   | 3.5471354 | -3.3450869 | -0.3979541 |
| Mo   | -0.4012204 | 3.3488575 | -3.5468906 |
| Mo   | 3.3844164 | -2.5488316 | 2.6018892 |
| P    | 1.0889467 | 3.6331887 | -1.5706442 |
| P    | 2.0024682 | -1.6213776 | 0.7637327 |
| P    | 0.7619284 | 1.6214724 | -2.0028695 |
| P    | 1.5701180 | -3.6330257 | 1.0912400 |
| O    | 5.1673093 | -0.6981984 | -0.8383515 |
| O    | 3.2176465 | 5.6195613 | -3.5929263 |
| O    | -0.8404819 | 0.6983454 | -5.1680202 |
| O    | 3.5892965 | -5.6184792 | 3.2225661 |
| O    | 4.5471804 | 2.5594789 | -0.9271959 |
| O    | 1.7730343 | -2.4117896 | -2.7999052 |
| O    | -2.8018371 | 2.4091813 | -1.7722814 |
| C    | -0.6255143 | 1.6378914 | -4.5385791 |
| O    | 0.9249576 | -2.5558651 | 4.5491336 |
| C    | 4.5381644 | -1.6378618 | -0.6230551 |
| C    | 2.9458249 | 4.5076192 | -3.4889003 |
| C    | 3.4860718 | -4.5066327 | 2.9500227 |
| C    | 4.7200817 | -4.5800496 | -1.9971708 |
| H    | 4.8834164 | -4.1995732 | -2.9919944 |
| C    | 3.7916387 | 2.5615371 | -1.7866411 |
| C    | 5.5871949 | -1.5528611 | 2.4162018 |
| H    | 6.2264063 | -1.6425344 | 1.5552301 |
| C    | -0.8946464 | 4.4326544 | -5.5962265 |
| Atoms | X          | Y          | Z          |
|-------|------------|------------|------------|
| H     | -0.8998763 | 3.9011159  | -6.5342884 |
| C     | 2.4134991  | 1.5527426  | -5.5883111 |
| H     | 1.5525284  | 1.6420650  | -6.2275751 |
| C     | 0.2005266  | 5.1481513  | -5.0527388 |
| H     | 1.1698395  | 5.2609093  | -5.5062474 |
| C     | 5.5973007  | -4.324787 | -0.8882645 |
| H     | 6.5352593  | -3.9007497 | -0.8922719 |
| C     | 1.7846670  | -2.5589386 | 3.7939012  |
| C     | 3.6221231  | -5.3890943 | -1.5734809 |
| H     | 2.7975352  | -5.7096299 | -2.1891720 |
| C     | 4.4693934  | 1.9261662  | -4.626512  |
| H     | 5.4426315  | 2.3396107  | -4.4191899 |
| C     | 4.6254699  | -1.9256650 | 4.4721794  |
| H     | 4.4178934  | -2.3388974 | 5.4454816  |
| C     | 5.5615878  | -2.4311339 | 3.5273841  |
| H     | 6.1759724  | -3.3078819 | 3.6562023  |
| C     | 5.0524554  | -5.1479586 | 0.2062476  |
| H     | 5.5046525  | -5.2604847 | 1.1761968  |
| C     | 2.6551916  | 0.5055853  | -4.6588659 |
| H     | 2.0036945  | -0.3288543 | -4.4583970 |
| C     | 3.5246026  | 2.4311162  | -5.5630395 |
| H     | 3.6533443  | 3.3074293  | -6.177803  |
| C     | 2.3824016  | -2.7336958 | -1.8813418 |
| C     | -1.8837051 | 2.7319704  | -2.3818516 |
| C     | -2.0023352 | 4.5798355  | -4.7174386 |
| H     | -2.9973091 | 4.1991985  | -4.8794676 |
| C     | -1.5772667 | 5.3886860  | -3.6198585 |
| H     | -2.1918734 | 5.7089135  | -2.7943398 |
| C     | 3.9228171  | 0.7314743  | -4.0671172 |
| H     | 4.4067880  | 0.0991282  | -3.3411295 |
| C     | 4.0664753  | -0.7307636 | 3.9255293  |
| H     | 3.3407856  | -0.0980541 | 4.4094673  |
| C     | -0.2203914 | 5.7339535  | -3.8278432 |
| H     | 0.3801342  | 6.3585036  | -3.1881463 |
| C     | 3.8283023  | -5.7341504 | -0.2162818 |
| H     | 3.1879172  | -6.3587595 | 0.3834486  |
| C     | 4.6582400  | -0.5052522 | 2.6578521  |
H  4.4580764  0.3292118  2.0062750
Ag  0.0000315 -0.0002792  0.0000874
Mo -2.6002779  2.5508524  3.3840327
Mo -3.5455346 -3.3464935  0.4002374
Mo  0.3996163  3.3474759  3.5448068
Mo -3.3830817 -2.5521613 -2.6002138
P  -1.0905294  3.6338697  1.5682606
P  -2.0017261 -1.6227467 -0.7625935
P  -0.7626954  1.6225720  2.0018443
P  -1.5683170 -3.6343894 -1.0886948
O  -5.1668448 -0.7000165  0.839295
O  -3.2203125  5.6206422  3.5890415
O   0.8398085  0.7022852  5.1678292
O  -3.5864306 -5.6223319 -3.2187859
O  -4.5482802  2.5582722  0.9252210
O  -1.7719483 -2.4107832  2.8016163
O   2.8007887  2.4100710  1.7710358
C   0.6245125  1.6413016  4.5377101
O  -0.9234194 -2.5590423 -4.5471922
C  -4.5373519 -1.6395859  0.6242290
C  -2.9480072  4.5087491  3.4857825
C  -3.4837397 -4.5102418 -2.9470410
C  -4.7179371 -4.5809385  2.0022446
H  -4.8815112 -4.1998534  2.9947971
C  -3.7927840  2.5612090  1.7847041
C  -5.5863421 -1.5570553 -2.4153371
H  -6.2255621 -1.6463813 -1.5543315
C   0.8923790  4.4369184  5.5934379
H   0.8977048  3.9060622  6.5318856
C  -2.4145783  1.5553519  5.5871143
H  -1.5537175  1.6453216  6.2264314
C  -0.2029940  5.1516328  5.0493209
H  -1.1723842  5.2643925  5.5026632
C  -5.5951836 -4.4345929  0.8911995
H  -6.5334155 -3.9033392  0.8947927
C  -1.7832078 -2.5621841 -3.7920547
C  -3.6195555 -5.3897293  1.5771756
H  -2.7948408  -5.7094245  2.1931363
C  -4.4705464   1.9274644  4.6251153
H  -5.4439194   2.3404404  4.4173523
C  -4.6243493  -1.9309090 -4.4709984
H  -4.4165443  -2.3447380 -5.4439994
C  -5.5602861  -2.4361260 -3.5258809
H  -6.1742730  -3.3130447 -3.6540930
C  -5.0499135  -5.1505548  0.2027911
H  -5.5020408  -5.2640379 -1.1726636
C  -2.6558456   0.5075391  4.6582890
H  -2.0040700  -0.3268210  4.4583919
C  -3.5259911   2.4333145  5.5612503
H  -3.6550846   3.099504  6.1754584
C  -2.3811211  -2.7335681  1.8832304
C  1.8824623   2.7343980  2.3803150
C  2.0001149   4.5838967  4.7146770
H  2.9952178   4.2037645  4.870989
C  1.5748710   5.3918099  3.6164781
H  2.1894596   5.7116964  2.7908106
C  -3.9235231   0.7326266  4.0663422
H  -4.4072397   0.0996627  3.3407235
C  -4.0659257  -0.7353569 -3.9251811
H  -3.3404976  -0.1026621 -4.4095407
C  0.2178434   5.7367178  3.8240562
H  -0.3828427   6.3605895  3.1838458
C  -3.8254766  -5.7358293  0.2202014
H  -3.1847460  -6.3605390 -0.3790573
C  -4.6578516  -0.5092106  2.6576968
H  -4.4580712   0.3257932 -2.0066994
Table S11. Cartesian coordinates of the gas-phase optimized geometry of \([\text{Cp(CO)}_2\text{Mo}_2(\mu-P_2)]_2\text{Ag}\)\(^+\) at the RI-B3LYP/def2-TZVP level of theory. (E = -5125.363715813 a.u.).

| Atom | x    | y    | z    |
|------|------|------|------|
| Mo   | 2.3164711 | 3.3395134 | -1.8833459 |
| Mo   | 3.6185776 | -2.8861013 | 1.0701075 |
| Mo   | -0.1963182 | 2.6467687 | -3.5973463 |
| Mo   | 2.6896647 | -1.5509191 | 3.7270089 |
| P    | 0.0126015 | 2.4741225 | -1.1311430 |
| P    | 1.7179217 | -1.2132446 | 1.4872511 |
| P    | 1.2783153 | 1.1025696 | -2.1595123 |
| P    | 1.3869776 | -3.1631022 | 2.1566106 |
| O    | 4.9014448 | -0.1377997 | 0.2610138 |
| O    | 0.8004704 | 5.9919356 | -1.1422475 |
| O    | 1.8383548 | 1.5003897 | -5.7017721 |
| O    | 2.9072860 | -4.3788368 | 5.0760173 |
| O    | 2.7612735 | 2.9009014 | 1.1819360 |
| O    | 2.2060494 | -2.9642801 | -1.7284965 |
| O    | -1.7158629 | -0.0807917 | -3.7993283 |
| C    | 1.1310931 | 1.9029613 | -4.8929562 |
| O    | -0.1785674 | -1.3071133 | 4.9788403 |
| C    | 4.3903729 | -1.1200686 | 0.5787556 |
| C    | 1.3041319 | 4.9977488 | -1.4150167 |
| C    | 2.8040488 | -3.3679767 | 4.5411786 |
| C    | 5.2959135 | -4.1942694 | 0.1069232 |
| H    | 5.6508631 | -4.0087830 | -0.8933668 |
| C    | 2.5535356 | 3.0450403 | 0.0621835 |
| C    | 4.8203871 | -0.4078366 | 3.7789945 |
| H    | 5.6374066 | -0.6170438 | 3.1103106 |
| C    | -0.6524694 | 4.2664838 | -5.2727993 |
| H    | -0.0223723 | 4.3845264 | -6.1398395 |
| C    | 3.6057055 | 3.7319405 | -3.8963333 |
| H    | 3.1816810 | 3.8192117 | -4.8840400 |
| C    | -0.5500208 | 4.9987997 | -4.0647020 |
| H    | 0.1656252 | 5.7748710 | -3.8548840 |
| C    | 5.8446517 | -3.6598592 | 1.3054139 |
| H    | 6.6805123 | -2.9817045 | 1.3676955 |
| C    | 0.8456611 | -1.4304738 | 4.4841792 |
| Atom | X       | Y       | Z       |
|------|---------|---------|---------|
| C    | 4.206478| -5.0826350| 0.4769142 |
| C    | 3.6433974 | -5.6721254 | -0.1997394 |
| C    | 4.4572578 | 4.2691855 | -1.8315798 |
| H    | 4.8023876 | 4.8379232 | -0.9837571 |
| C    | 3.4450252 | -0.4281510 | 5.6237677 |
| H    | 3.0483119 | -0.6436442 | 6.6022081 |
| C    | 4.5918345 | -1.0286936 | 5.0320965 |
| H    | 5.2046441 | -1.7930391 | 5.4823233 |
| C    | 5.1407712 | -4.2161809 | 2.4020780 |
| H    | 5.3500992 | -4.0406494 | 3.4429604 |
| C    | 4.0846761 | 2.5361213 | -3.2990129 |
| H    | 4.0797929 | 1.5578656 | -3.7498165 |
| C    | 3.8294037 | 4.7970413 | -2.9921353 |
| H    | 3.5971337 | 5.8355316 | -3.1659467 |
| C    | 2.6912939 | -2.9019599 | -0.6920455 |
| C    | -1.1272494 | 0.8992751 | -3.6959585 |
| C    | -1.7606870 | 3.3829938 | -5.1652361 |
| H    | -2.1288548 | 2.7309010 | -5.9402055 |
| C    | -2.3418987 | 3.5723053 | -3.8733387 |
| H    | -3.2155143 | 3.0714252 | -3.4886521 |
| C    | 4.6164464 | 2.8622192 | -2.0268095 |
| H    | 5.0848843 | 2.1734765 | -1.3423991 |
| C    | 2.9649932 | 0.5645295 | 4.7167523 |
| H    | 2.1268938 | 1.2217406 | 4.8830188 |
| C    | -1.5900992 | 4.5650553 | -3.1984878 |
| H    | -1.7916224 | 4.9503876 | -2.2129566 |
| C    | 4.1451269 | -5.0919574 | 1.888162 |
| H    | 3.4611164 | -5.6834399 | 2.4740919 |
| C    | 3.8115008 | 0.5727481 | 3.5811772 |
| H    | 3.7188712 | 1.2286261 | 2.7324114 |
| Ag   | -0.0024272 | -0.0064474 | 0.035047 |
| Mo   | -4.5741200 | 0.2938296 | 1.2868961 |
| Mo   | -4.1081857 | -2.5363891 | 0.0716322 |
| P    | -2.4204544 | -0.7669317 | 0.3773915 |
| P    | -3.0881965 | -1.4837940 | 2.2206793 |
| O    | -4.3856261 | 1.4581428 | -1.6211431 |
| O    | -5.8200222 | -3.6836886 | 2.4415784 |
| Atoms | Ox | X1  | Y1  | Z1   | Ox | X2  | Y2  | Z2   |
|-------|----|-----|-----|------|----|-----|-----|------|
| O     | -2.4856664 | 2.4473351 | 2.2007318 |
| O     | -1.9443093 | -4.7060413 | 0.7509689 |
| C     | -4.4152561 | 0.9961427 | -0.5660508 |
| C     | -5.1692092 | -3.2328212 | 1.6091983 |
| C     | -6.2183209 | 1.7878680 | 2.0067259 |
| H     | -6.1339424 | 2.8474451 | 1.8300668 |
| C     | -5.5265993 | -1.8716627 | -1.7708356 |
| H     | -6.0979010 | -0.9598861 | -1.7979800 |
| C     | -6.8744916 | 0.8461691 | 1.1668130 |
| H     | -7.3597602 | 1.0686396 | 0.2300479 |
| C     | -2.7144581 | -3.8846576 | 0.5475994 |
| C     | -5.7624629 | 1.0862362 | 3.1642116 |
| H     | -5.2527861 | 1.5218611 | 4.0083304 |
| C     | -4.9400925 | -4.0758790 | -1.4668890 |
| H     | -4.9954229 | -5.1275584 | -1.2387826 |
| C     | -5.9723110 | -3.1173159 | -1.2631769 |
| H     | -6.9419517 | -3.3180717 | -0.8366341 |
| C     | -6.8290034 | -0.4204344 | 1.8003109 |
| H     | -7.2783358 | -1.3272804 | 1.4344128 |
| C     | -3.2130080 | 1.6358485 | 1.8458969 |
| C     | -3.8501159 | -3.4042815 | -2.090053 |
| H     | -2.9258326 | -3.8583949 | -2.4175708 |
| C     | -6.1364926 | -0.2722308 | 3.0333216 |
| H     | -5.9573485 | -1.0513443 | 3.7552109 |
| C     | -4.2127697 | -2.0473694 | -2.2823489 |
| H     | -3.6068910 | -1.2918105 | -2.7529039 |
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