Structural and coordination properties of 1,2-bis(cyclopropyl)-3,4-bis(2,4,6-tri-*tert*-butylphenyl)-3,4-diphosphinidenecyclobutene prepared by dehydrogenative homocoupling of 3-cyclopropyl-1-(2,4,6-tri-*tert*-butylphenyl)-1-phosphaallene

Shigekazu Ito,* Matthias Freytag and Masaaki Yoshifuji*

Received 21st July 2005, Accepted 28th September 2005
First published as an Advance Article on the web 31st October 2005
DOI: 10.1039/b510432g

According to a protocol for the synthesis of phosphaallenes we recently established, 3-cyclopropyl-1-(2,4,6-tri-*tert*-butylphenyl)-1-phosphallene was obtained from (*Z*)-2-bromo-3-cyclopropyl-1-(2,4,6-tri-*tert*-butylphenyl)-1-phosphapropene. A novel bidentate ligand, 1,2-bis(cyclopropyl)-3,4-bis(2,4,6-tri-*tert*-butylphenyl)-3,4-diphosphinidenecyclobutene, was prepared by oxidative homocoupling of 3-cyclopropyl-1-(2,4,6-tri-*tert*-butylphenyl)-1-phosphaallene in the presence of butyllithium, together with the generation of hydrogen gas. The 1,2-bis(cyclopropyl)-3,4-diphosphinidenecyclobutene was allowed to react with (tht)AuCl (tht = tetrahydrothiophene) to afford the corresponding digold(I) complex of a six-membered metallacycle containing the P=C–C=P skeleton. The molecular structures of the 2-bromo-3-cyclopropyl-1-phosphapropene, 1,2-bis(cyclopropyl)-3,4-diphosphinidenecyclobutene and the digold complex were unambiguously determined by X-ray crystallography and are discussed from the point of view of the cyclopropyl conjugation.

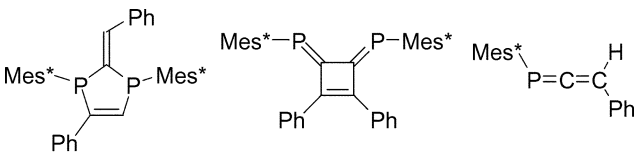
Introduction

As cumulene compounds are widely employed for organic synthesis,[1] 1-phosphaallenes [–P=C=C<] are expected to be promising materials for the synthesis of novel phosphorus compounds.[2] We previously reported topochemical head-to-tail [2 + 2] dimerisation reactions in the solid state[3] and a formal [3 + 2] dimerisation affording a 1,4-diphosphafulvene (**1**),[4] suggesting the utility of 1-phosphaallenes in organic chemistry featuring phosphorus. These findings motivated us to develop the chemistry of phosphacumulene derivatives leading to material science.

In the study of the 1,4-diphosphafulvene **1**, we found the formation of 3,4-diphosphinidenecyclobutene **2** together with **1** from 3-phenyl-1-phosphaallene **3** in the presence of a base.[4] The 3,4-diphosphinidenecyclobutenes (DPCB) have been utilised as π-electron accepting ligands for synthetic catalysts,[5] and furthermore, attempts have been made to prepare DPCB derivatives involving molecular functionality.[6] Taking our previous results into account,[4] 1-phosphaallenes are expected to be versatile reagents for the synthesis of DPCB derivatives. We have established some practical synthetic procedures of 1-(2,4,6-tri-*tert*-butylphenyl)-1-phosphaallenes from 2-bromo-1-(2,4,6-tri-*tert*-butylphenyl)-1-phosphapropenes,[3,4,7] which encouraged us to utilise 1-phosphaallenes for the preparation of novel organic compounds containing phosphorus.

On the other hand, we previously found that the P=C double bond conjugates with a cyclopropyl group on the basis of the structures of 2-cyclopropyl-1-phosphaethenes,[8] which prompted us to investigate properties of 3-cyclopropyl-1-phosphaallenes.

Department of Chemistry, Graduate School of Science, Tohoku University, Aoba, Sendai, 980-8578, Japan. E-mail: yoshifj@mail.tains.tohoku.ac.jp, ito@synchem.chem.tohoku.ac.jp; Fax: +81-22-795-6562



1 **2** **3**

Mes* = 2,4,6-*t*-$Bu_3C_6H_2$

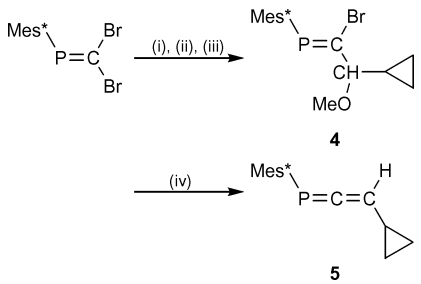
The synthesis of a 3-cyclopropyl-1-phosphaallene was successful, and therefore, we proceeded to investigate the chemistry of phosphacumulenes bearing a cyclopropyl group. In the course of our studies of 3-cyclopropyl-1-phosphaallenes, we found an oxidative homocoupling affording a novel 3,4-diphosphinidenecyclobutene. In this paper we describe the generation, isolation and molecular structure of a 1,2-bis(cyclopropyl)-3,4-diphosphinidenecyclobutene prepared simply by mixing a 3-cyclopropyl-1-phosphaallene and butyllithium. Moreover, we report that the two sp^2-hybridised phosphorus atoms of the 3,4-diphosphinidenecyclobutene coordinate on two gold(I) atoms, respectively, affording a novel digold(I) complex, stabilised by an intramolecular gold–gold contact. This observation is in contrast to the well-established coordination chemistry of DPCB compounds, which in most cases act as chelating ligands to one single metal centre, forming a five-membered metallacycle. The effects of cyclopropyl conjugation are discussed, based on the molecular structures.

Results and discussion

Preparation of 3-cyclopropyl-1-(2,4,6-tri-*tert*-butylphenyl)-1-phosphaallene

2,2-Dibromo-1-(2,4,6-tri-*tert*-butylphenyl)-1-phosphaethene [Mes*P=CBr_2] was allowed to react successively with butyllithium,

cyclopropanecarbaldehyde, and iodomethane[4] to afford 2-bromo-3-cyclopropyl-1-phosphapropene **4** in 63% yield (Scheme 1). The structure of **4** was confirmed by X-ray crystallography as displayed in Fig. 1. The cyclopropyl ring forms an equilateral triangle, indicating almost no conjugative interaction with any π-functional groups. On the other hand, the cyclopropyl triangle of 2-cyclopropyl-1-phosphaethene is isosceles, indicating the cyclopropyl conjugation.[8] Compound **4** was allowed to react with *tert*-butyllithium to afford 3-cyclopropyl-1-phosphaallene **5** in 63% yield. Although the X-ray structure determination of **5** was incomplete,[9] it indicated that the P=C=C skeleton and the cyclopropyl group show a conformation of *s-trans* type and that the cyclopropyl ring is almost perpendicular to the adjacent C=C π-system.[10]



Scheme 1 *Reagents and conditions*: (i) *n*-BuLi, THF, −100 °C; (ii) *c*-PrCHO, −100 °C to rt; (iii) MeI; (iv) *t*-BuLi, THF, −100 °C to rt.

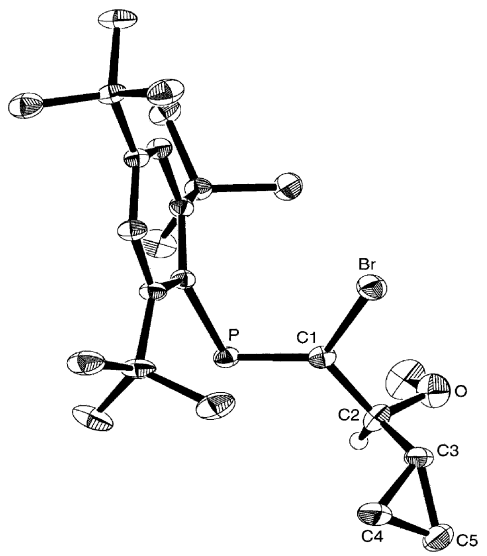
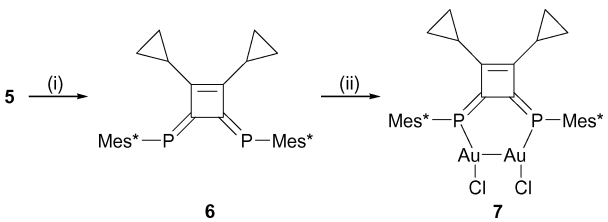


Fig. 1 Molecular structure of **4**. Hydrogen atoms except for the CH hydrogen are omitted for clarity. Selected bond lengths (Å) and angles (°): Br–C1 1.901(3), P–C1 1.671(3), P–C_{Mes*} 1.841(3), C1–C2 1.533(4), C2–O 1.404(5), O–Me 1.411(5), C2–C3 1.488(5), C3–C4 1.495(5), C3–C5 1.492(5), C4–C5 1.493(6); C1–P–C_{Mes*} 105.0(1), Br–C1–P 125.5(2), Br–C1–C2 114.6(2), P–C1–C2 120.0(2), C1–C2–C3 112.3(3), C2–C3–C4 118.8(3), C2–C3–C5 119.5(3), C4–C3–C5 60.0(3), C3–C4–C5 59.9(3), C3–C5–C4 60.1(2).

Preparation and structure of 1,2-bis(cyclopropyl)-3,4-bis(2,4,6-tri-*tert*-butylphenyl)-3,4-diphosphinidenecyclobutene

3-Cyclopropyl-1-phosphaallene **5** was allowed to react with 0.5 equiv. of butyllithium in THF. In the reaction mixture, 3,4-diphosphinidenecyclobutene **6** was observed together with **5** by ^{31}P NMR spectroscopy in a 2 : 1 ratio (Scheme 2). Compound **6** was recrystallised from hexane to obtain a suitable single crystal for X-ray structure determination. In this reaction, no 1,4-diphosphafulvene derivative was observed in the reaction mixture, which shows a sharp contrast to the reactions of 3-phenyl-1-phosphaallene **3**.[4,11] Generation of hydrogen was observed in the formation of **6**, which is similar to the formation of phenanthrenequinone from benzil in the presence of C_8K.[12] No oxidative reagent is required in the preparation of **6** from **5**, which leads to the establishment of a concise method for the synthesis of 3,4-diphosphinidenecyclobutene derivatives. We examined the reaction of **5** with 1 equiv. of butyllithium, but the yield of **6** was even lower than in the case using 0.5 equiv. of butyllithium, as mentioned above. The reaction mechanism of the formation of **6** from **5** in the presence of butyllithium might include a catalytic dehydrogenation under basic conditions, and attempts to clarify the details are underway. The melting point of **6** is considerably high, which is similar to the property of hexacyclopropylbenzene (266–270 °C).[13] The molecular structure of **6** was confirmed by X-ray crystallography as shown in Fig. 2. The two cyclopropyl groups are perpendicular to the nearly planar cyclobutene ring, which indicates the cyclopropyl conjugation.[4,10] However, the cyclopropyl groups of **6** do not take an exactly bisected conformation to the cyclobutene ring, while the molecule shows a C_2 symmetric structure. On the other hand, the cyclopropyl groups in hexacyclopropylbenzene are reported not to take a perpendicular alignment to the benzene ring, probably due to minimisation of the steric congestion.[13] The



Scheme 2 *Reagents and conditions*: (i) *n*-BuLi, THF, −78 °C to rt; (ii) (tht)AuCl, CH_2Cl_2, rt.

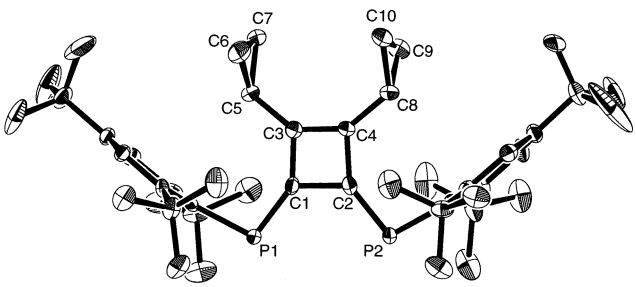


Fig. 2 Molecular structure of **6**. Hydrogen atoms are omitted for clarity. Selected bond lengths (Å) and angles (°): P1–C1 1.674(6), P1–C_{Mes*} 1.851(5), P2–C2 1.682(6), P2–C_{Mes*} 1.857(6), C1–C2 1.513(7), C1–C3 1.472(8), C2–C4 1.487(8), C3–C4 1.393(8), C3–C5 1.484(8), C5–C6 1.50(1), C5–C7 1.486(9), C6–C7 1.43(1), C4–C8 1.478(8), C8–C9 1.510(10), C8–C10 1.505(9), C9–C10 1.45(1); C1–P1–C_{Mes*} 100.2(2), C2–P2–C_{Mes*} 100.5(3), C2–C1–C3 87.7(4), C1–C2–C4 87.6(4), C1–C3–C4 92.9(5), C2–C4–C3 91.7(5), C4–C3–C5 139.2(6), C1–C3–C5 127.8(5), C3–C5–C6 120.7(7), C3–C5–C7 124.3(5), C6–C5–C7 57.4(5), C5–C6–C7 60.9(5), C5–C7–C6 61.7(5), C4–C8–C9 121.6(6), C4–C8–C10 123.8(5), C9–C8–C10 57.7(5), C8–C9–C10 61.0(5), C8–C10–C9 61.3(5).

proximal bonds of the cyclopropyl groups of **6** are longer than the distal bonds, indicating the cyclopropyl conjugation.

Preparation and structure of the digold(I) complex of the 1,2-bis(cyclopropyl)-3,4-diphosphinidenecyclobutene

3,4-Diphosphinidenecyclobutene **6** is expected to function as a P_2-ligand for metal complex formation, and we chose the gold(I) reagent (tht)AuCl (tht = tetrahydrothiophene) to obtain a novel complex bearing **6** in this study. Compound **6** was allowed to react with (tht)AuCl in dichloromethane at room temperature and, after the workup procedure, the digold(I) complex **7** was obtained in 39% yield. Complex **7** is slightly unstable in solution as could be observed by the formation of a gold mirror on the used glassware. No monogold(I) complex was observed in the reaction mixture of **6** and (tht)AuCl. The molecular structure of **7** was determined by X-ray crystallography, and Fig. 3 displays its ORTEP drawing. In spite of steric congestion and the rigid skeleton around the phosphorus atoms, two gold atoms are included in the molecule. The molecule shows C_2 symmetry, and the cyclopropyl groups take an "alternating" conformation, probably to minimise the steric congestion. Indeed, the cyclopropyl groups of **7** turn up and down probably due to the distortion caused by the coordination of two gold atoms. The torsion angle of Θ(P1–C1–C2–P2) (26°) is larger than that of **6** (0.5°). The six-membered metallacycle disturbs the planar 3,4-diphosphinidenecyclobutene skeleton [Θ(Au2–Au1–P1–C1) = 50.8°; Θ(Au1–Au2–P2–C2) = 44.6°]. The Au1–Au2 distance is close to that of [$Ph_2P(CH_2)_2PPh_2$][AuCl]$_2$ [**8**: 3.05(1) Å],[14] indicating an aurophilic attraction.[15] The Au–P and Au–Cl distances are comparable to those of **8**.[14]

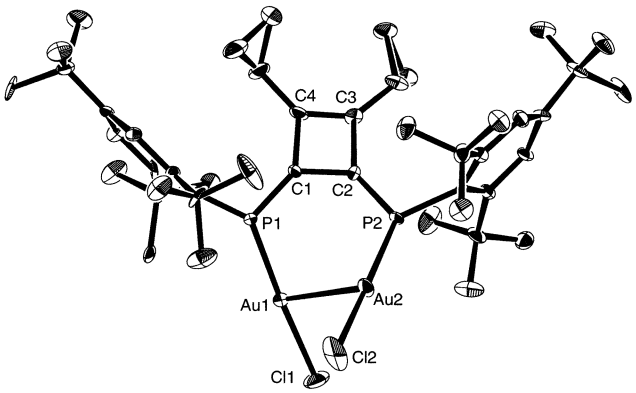


Fig. 3 Molecular structure of **7**. Hydrogen atoms are omitted for clarity. Selected bond lengths (Å) and angles (°): Au1–Au2 3.0096(5), Au1–Cl1 2.293(3), Au2–Cl2 2.280(3), Au1–P1 2.223(3), Au2–P2 2.220(3), P1–C1 1.662(9), P1–C_{Mes*} 1.834(9), P2–C2 1.670(9), P2–C_{Mes*} 1.816(9), C1–C2 1.49(1), C1–C4 1.48(1), C2–C3 1.48(1), C3–C4 1.40(1); Au2–Au1–Cl1 89.64(7), Au2–Au1–P1 91.27(6), Cl1–Au1–P1 174.7(1), Au1–Au2–Cl2 94.58(9), Au1–Au2–P2 89.43(6), Cl2–Au2–P2 174.1(1), Au1–P1–C1 116.1(3), Au1–P1–C_{Mes*} 136.7(3), C1–P1–C_{Mes*} 107.0(4), Au2–P2–C2 119.3(3), Au2–P2–C_{Mes*} 133.7(3), C2–P2–C_{Mes*} 105.5(4), P1–C1–C2 131.7(3), P1–C1–P4 140.6(7), C2–C1–C4 87.7(3), P2–C2–C1 131.7(3), P2–C2–C3 138.9(7), C1–C2–C3 88.1(7), C1–C4–C3 91.7(8).

Conclusion

We have demonstrated the formation of 1,2-bis(cyclopropyl)-3,4-diphosphinidenecyclobutene **6** from 3-cyclopropyl-1-phosphaallene **5** in the presence of a base, together with the generation of hydrogen gas. The molecular structure of **6** revealed an effect of the cyclopropyl conjugation with the DPCB skeleton. Two gold atoms can coordinate on the two phosphorus atoms of **6**, indicating that the aurophilic interaction overcomes the steric congestion caused by the bulky Mes* groups and the rigid DPCB skeleton. Further investigation on properties of **4**–**7** is in progress to develop novel functional phosphorus compounds.

Experimental

Preparation of 4

To a solution of 2,2-dibromo-1-(2,4,6-tri-*tert*-butylphenyl)-1-phosphaethene (1.00 g, 2.23 mmol) in THF (20 mL) was added butyllithium (3.30 mmol, 1.6 M solution in hexane, 1 M = 1 mol dm^{-3}) at −100 °C. After being stirred for 10 min, the mixture was then treated with cyclopropanecarbaldehyde (2.23 mmol) and allowed to warm to room temperature. An excess equiv. of iodomethane (4.5 mmol) was added and the reaction mixture was stirred for 15 min. The solvent and volatile materials were removed *in vacuo*, and the residue was extracted with hexane. Silica-gel column chromatography (hexane/EtOAc = 20 : 1) of the hexane extracts gave 750 mg of **4** (yield: 63%), mp 76–78 °C; $^{31}P\{^1H\}$ NMR (162 MHz, $CDCl_3$) δ 259.6; ^1H NMR (400 MHz, $CDCl_3$) δ 7.46 (2H, s, arom), 3.37 (3H, s, OMe), 3.31 (1H, m, C*H*–OMe), 1.55 (9H, s, *o*-*t*Bu), 1.53 (9H, s, *o*-*t*Bu), 1.38 (9H, s, *p*-*t*Bu), 1.35 (1H, m, CH), 0.73 (1H, m, C*H*H), 0.59 (2H, m, CH*H*), 0.38 (1H, m, C*H*H); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 166.4 (d, $^1J_{PC}$ = 63.8 Hz, P=C), 153.8 (d, $^2J_{PC}$ = 2.5 Hz, *o*-arom), 153.6 (d, $^2J_{PC}$ = 2.5 Hz, *o*-arom), 151.3 (s, *p*-arom), 136.6 (d, $^1J_{PC}$ = 54.3 Hz, *ipso*-arom), 122.8 (s, *m*-arom), 122.5 (s, *m*-arom), 91.4 (d, $^2J_{PC}$ = 40.0 Hz, CH), 57.3 (s, OMe), 38.3 (s, *o*-*C*Me$_3$), 38.2 (s, *o*-*C*Me$_3$), 35.5 (s, *p*-*C*Me$_3$), 33.4 (d, $^4J_{PC}$ = 6.9 Hz, *o*-C*Me*$_3$), 33.2 (d, $^4J_{PC}$ = 6.5 Hz, *o*-C*Me*$_3$), 31.8 (s, *p*-C*Me*$_3$), 16.9 (d, $^3J_{PC}$ = 11.6 Hz, CH-cyclopropyl), 5.4 (s, CH_2), 4.2 (s, CH_2); HRMS: calc. for $C_{24}H_{38}BrOP$ + Na: 475.1736; found: 475.1738.

Preparation of 5

To a solution of 635 mg (1.4 mmol) of **4** in THF (30 mL) was added *tert*-butyllithium (1.4 mmol) at −100 °C. After being stirred for 10 min, the reaction mixture was allowed to warm to room temperature and the volatile materials were removed *in vacuo*. The residue was extracted with hexane and purified by column chromatography (hexane) to give 300 mg of **5** (yield: 63%), mp 75–77 °C; $^{31}P\{^1H\}$ NMR (162 MHz, $CDCl_3$) δ 70.6; ^1H NMR (400 MHz, $CDCl_3$) δ 7.47 (2H, s, arom), 5.33 (1H, dd, $^3J_{PH}$ = 26.2 Hz, $^3J_{HH}$ = 5.6 Hz, C=CH), 1.67 (18H, s, *o*-*t*Bu), 1.49 (1H, m, CH), 1.38 (9H, s, *p*-*t*Bu), 0.80 (2H, m, C*H*H), 0.42 (2H, m, CH*H*); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 239.1 (d, $^1J_{PC}$ = 24.0 Hz, P=C), 153.9 (d, $^2J_{PC}$ = 3.4 Hz, *o*-arom), 149.9 (s, *p*-arom), 133.1 (d, $^1J_{PC}$ = 64.3 Hz, *ipso*-arom), 122.4 (s, *m*-arom), 116.6 (d, $^2J_{PC}$ = 15.2 Hz, C=*C*H), 38.6 (s, *o*-*C*Me$_3$), 35.4 (s, *p*-*C*Me$_3$), 33.6 (d, $^4J_{PC}$ = 7.4 Hz, *o*-C*Me*$_3$), 31.8 (s, *p*-C*Me*$_3$), 10.5 (d, $^3J_{PC}$ = 15.1 Hz, CH), 7.9 (d, $^3J_{PC}$ = 2.8 Hz, CH_2), 7.5 (s, CH_2); HRMS: calc. for $C_{23}H_{35}P$ + Na: 365.2369; found: 365.2366.

Preparation of 6

To a solution of 300 mg (0.88 mmol) of **5** in THF (12 mL) was added butyllithium (0.44 mmol, 1.6 M solution in hexane)

at −78 °C. After being stirred for 10 min, the reaction mixture was allowed to warm to room temperature and the volatile materials were removed *in vacuo*. The residue was extracted with hexane and evaporated *in vacuo*. Recrystallisation from hexane gave 48 mg of **6** (yield: 16%), mp 312–315 °C; $^{31}P\{^1H\}$ NMR (162 MHz, $CDCl_3$) δ 147.0; ^1H NMR (400 MHz, $CDCl_3$) δ 7.33 (4H, s, arom), 1.57 (36H, s, *o*-*t*Bu), 1.28 (18H, s, *p*-*t*Bu), 0.22 (4H, m, C*H*H), 0.11 (4H, m, CH*H*), −0.31 (2H, m, CH); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 181.3 (dd, $^1J_{PC}$ = 19.8 Hz, $^2J_{PC}$ = 5.4 Hz, P=C), 162.1 (dd, $^2J_{PC}$ = 7.8 Hz, $^3J_{PC}$ = 5.8 Hz, C=C), 155.9 (s, *o*-arom), 149.9 (s, *p*-arom), 136.5 (dd, $^1J_{PC}$ = 29.6 Hz, $^4J_{PC}$ = 25.4 Hz, *ipso*-arom), 121.3 (s, *m*-arom), 38.5 (s, *o*-*C*Me$_3$), 35.3 (s, *p*-*C*Me$_3$), 33.5 (pt, ($^4J_{PC}$ + $^7J_{PC}$)/2 = 3.5 Hz, *o*-C*Me*$_3$), 31.7 (s, *p*-C*Me*$_3$), 9.8 (s, CH_2), 8.4 (s, CH); HRMS: calc. for $C_{46}H_{68}P_2$ + Na: 705.4688; found: 705.4690.

Preparation of 7

A mixture of 25 mg (0.036 mmol) of **6** and 23.5 mg (0.073 mmol) of (tht)AuCl in 8 mL of CH_2Cl_2 was stirred for 1 h, then the volatile materials were removed *in vacuo*. Recrystallisation from a mixture of CH_2Cl_2 and hexane gave 16 mg of **7** (yield: 39%). The reaction and the recrystallisation were carried out in the dark, mp 238–239 °C; $^{31}P\{^1H\}$ NMR (162 MHz, $CDCl_3$) δ 113.7; ^1H NMR (400 MHz, $CDCl_3$) δ 7.48 (4H, s, arom), 1.71 (36H, s, *o*-*t*Bu), 1.28 (18H, s, *p*-*t*Bu), 0.39 (4H, m, C*H*H), 0.27 (4H, m, CH*H*), −0.37 (2H, m, CH); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 170.3 (dd, $^1J_{PC}$ = 38.2 Hz, $^2J_{PC}$ = 26.5 Hz, P=C), 163.8 (d, $^2J_{PC}$ = 26.1 Hz, C=C), 158.7 (s, *o*-arom), 154.7 (s, *p*-arom), 123.2 (pt, ($^3J_{PC}$ + $^6J_{PC}$)/2 = 4.9 Hz, *m*-arom), 39.3 (s, *o*-*C*Me$_3$), 35.7 (s, *p*-*C*Me$_3$), 34.6 (s, *o*-C*Me*$_3$), 31.5 (s, *p*-C*Me*$_3$), 10.8 (pt, ($^4J_{PC}$ + $^5J_{PC}$)/2 = 1.7 Hz, CH_2), 7.1 (s, CH) (*ipso*-arom could not be determined); HRMS: calc. for $C_{46}H_{68}Au_2Cl_2P_2$ + Na: 1169.3397; found: 1169.3403.

X-Ray crystallography

A Rigaku RAXIS-IV imaging plate detector with graphite-monochromated Mo-Kα radiation (λ = 0.71070 Å) was used. The structure was solved by direct methods (SIR92),[16] expanded using Fourier techniques (DIRDIF94),[17] and then refined by full-matrix least squares method. The data were corrected for Lorentz polarization effect. Structure solution, refinement, and graphical representation were carried out using the teXsan package.[18]

Crystal data. For **4**: $C_{24}H_{38}BrOP$, M = 453.44, triclinic, space group $P\bar{1}$ (no. 2), a = 9.9117(5), b = 13.2871(5), c = 9.7067(5) Å, α = 105.593(2), β = 96.440(4), γ = 90.888(3)°, V = 1222.1(1) Å^3, Z = 2, D_c = 1.232 g cm^{-3}, μ(Mo-Kα) = 1.763 mm^{-1}, T = 133 K, 9728 observed reflections, 5101 unique reflections (R_{int} = 0.024), $R1$ = 0.051 ($I > 2\sigma(I)$), R_w = 0.078 (all data).

For **6**: $C_{46}H_{68}P_2$, M = 682.99, monoclinic, space group $P2_1/n$ (no. 14), a = 15.9000(4), b = 24.1687(7), c = 11.3333(5) Å, β = 89.711(1)°, V = 4355.1(3) Å^3, Z = 4, D_c = 1.042 g cm^{-3}, μ(Mo-Kα) = 0.128 mm^{-1}, T = 140 K, 35515 observed reflections, 9885 unique reflections (R_{int} = 0.084), $R1$ = 0.097 ($I > 2\sigma(I)$), R_w = 0.108 (all data).

For **7**: $C_{46}H_{68}Au_2Cl_2P_2$, M = 1147.83, orthorhombic, space group $Pna2_1$ (no. 33), a = 19.4009(5), b = 15.0454(4), c = 15.8448(7) Å, V = 4625.0(3) Å^3, Z = 4, D_c = 1.648 g cm^{-3}, μ(Mo-Kα) = 6.573 mm^{-1}, T = 133 K, 36160 observed reflections, 5323 unique reflections (R_{int} = 0.065), $R1$ = 0.031 ($I > 2\sigma(I)$), R_w = 0.038 (all data).

CCDC reference numbers 272085–272087.

For crystallographic data in CIF or other electronic format see DOI: 10.1039/b510432g

Acknowledgements

This work was supported in part by Grants-in-Aid for Scientific Research (No. 13303039 and 14044012) from the Ministry of Education, Culture, Sports, Science and Technology of Japan. M. Freytag is grateful to the Japan Society for the Promotion of Science for Postdoctoral Fellowships for Foreign Researchers.

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