Stereospecific Transfer Reactions of Phenyl and Ethynyl Groups between Platinum(II) Atoms

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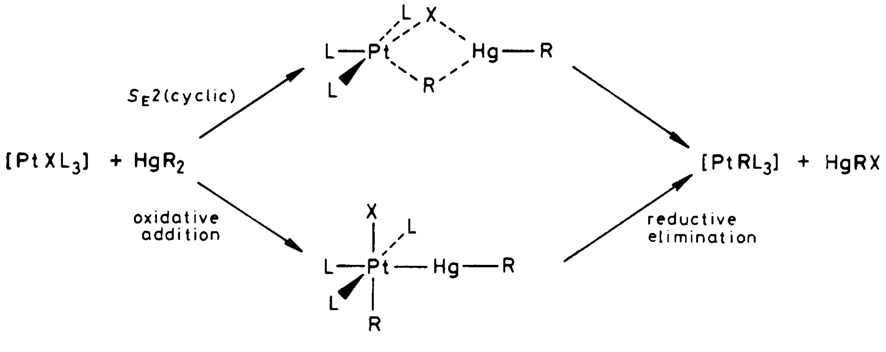
Treatment of *cis*-$[PtPh_2(CO)L]$, prepared from $[PtClPh(CO)L]$ ($L = PMePh_2$; Cl *trans* to L) and $HgPh_2$, with $HgCl_2$ results in the specific replacement by Cl of the phenyl group *trans* to L. The same phenyl is replaced when *cis*-$[PtPh_2(CO)L]$ and *cis*-$[PtCl_2(^{13}CO)L]$ react, forming $[PtClPh(CO)L]$ (Cl *trans* to L) and $[PtClPh(^{13}CO)L]$ (Ph *trans* to L). By contrast, the ethynyl complex *cis*-$[Pt(C{\equiv}CPh)_2(CO)L]$ reacts with *cis*-$[PtCl_2(CO)L]$ to form only one product, $[PtCl(C{\equiv}CPh)(CO)L]$ with Cl *trans* to L. The mixed bis(ethynyl) complex $[Pt(C{\equiv}CMe)(C{\equiv}CPh)(CO)L]$ ($C{\equiv}CMe$ *trans* to CO) reacts with *cis*-$[PtCl_2(^{13}CO)L']$ ($L' = PMe_2Ph$) to produce mainly $[PtCl(C{\equiv}CMe)(CO)L]$ and $[PtCl(C{\equiv}CPh)(^{13}CO)L']$ (both with Cl *trans* to phosphine) confirming that a chloride *trans* to CO is involved in this exchange. *cis*-$[Pt(C{\equiv}CPh)_2(^{13}CO)L']$ is also produced in this reaction, apparently *via* $[Pt(C{\equiv}CMe)(C{\equiv}CPh)(CO)L]$ and $[PtCl(C{\equiv}CPh)(^{13}CO)L']$. The mechanisms of these reactions are discussed.

It has become increasingly common in recent years to prepare organoplatinum compounds from organomercury [1-6] or organotin reagents.[7-10] The lower reactivity of these compounds compared to organolithium or Grignard reagents is frequently offset by high specificity and clean reaction conditions in replacing a halide by the organic group. The transfer mechanism has been the subject of much comment,[11] two routes in particular being singled out. The first is the S_E2-(cyclic) mechanism.[11-13] This involves simultaneous transfer of the two migrating groups, and is well documented for mercury.[14] The second is consecutive operation of oxidative addition and reductive elimination.[2,11,15,16] These are well known for square-planar d^8 complexes,[17] and examples involving mercury and tin are known.[15,18] The Scheme illustrates the two pathways.



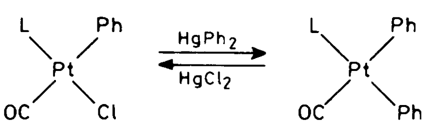
Scheme.

The transition state and intermediate shown in the Scheme are linked in as much as they can be regarded as extremes of a single process, the choice of description depending on the degree of Pt–Hg interaction at the time of transfer.[19,20] There are, however, other possible configurations for six-co-ordinate oxidative addition/reductive elimination intermediates, so this need not always be the case.

There have also been many reports in the literature of organic group transfers between two platinum atoms, two palladium atoms, or platinum and palladium. They include methyl,[11,20,21] phenyl,[11] and cyclopentadienyl.[22] There is little evidence for the mechanism of transfer, though the authors favour S_E2(cyclic) in most cases. We have examined a number of organic group transfers between mercury(II) and platinum complexes containing carbonyl and tertiary phosphine in *cis* arrangement.[20,22,23] In every case the group opposite tertiary phosphine was selectively replaced, presumably because of the higher bond-weakening *trans* influence [24] of PR_3 compared to CO. We report here an extension of these studies, including transfers of ethynyls between platinum atoms, where unexpected behaviour is observed.

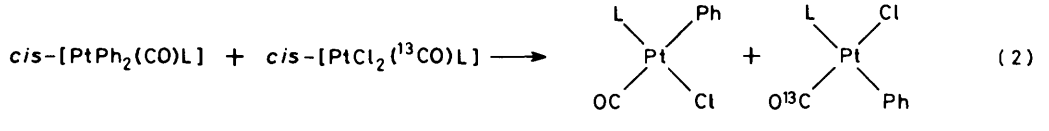
Results and Discussion

Phenyl Complexes.—*cis*-$[PtPh_2(CO)(PMePh_2)]$ has been synthesised previously by CO attack on *cis*-$[PtPh_2L_2]$ ($L = PMePh_2$).[25] We have prepared it by treating $[PtClPh(CO)L]$ (Cl *trans* to L) with $HgPh_2$. The reaction of $HgCl_2$ in chloroform regenerated the chlorophenylplatinum complex [equation (1); $L = PMePh_2$]. Both reactions were followed by



(1)

^{31}P n.m.r. spectroscopy in $CDCl_3$ solution. No other platinum-containing products were detected and these specific reactions, replacing the groups *trans* to $PMePh_2$, fit the pattern of other



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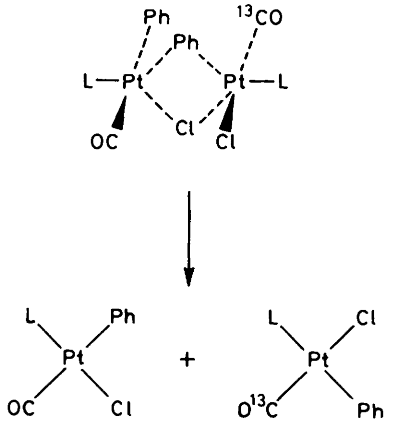

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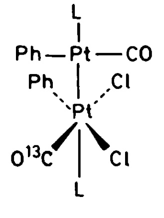
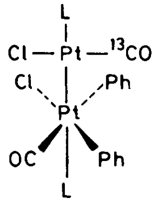


(A)



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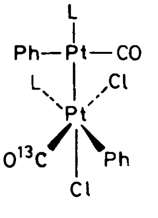
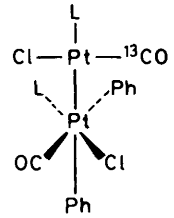


(C)



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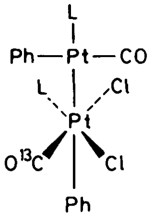

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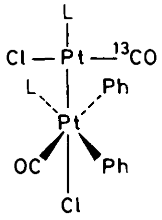
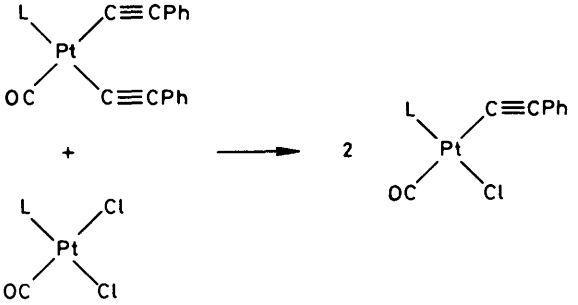


(F)

related substitutions,[20,22,23] and probably proceed *via* the S_E2(cyclic) type of mechanism.

A CDCl$_3$ solution of *cis*-[PtPh$_2$(CO)L] in CDCl$_3$ was treated with *cis*-[PtCl$_2$(^{13}CO)L], and the mixture similarly observed at ambient temperature by ^{31}P n.m.r. spectroscopy. After 5 min, no starting complex remained and the products consisted of a mixture of [PtClPh(CO)L] (Cl *trans* to L) and [PtClPh(^{13}CO)L] {Ph *trans* to L; this latter complex existing in equilibrium with its carbonyl insertion product, [Pt$_2$(^{13}COPh)$_2$(μ-Cl)$_2$L$_2$]},[23] equation (2). With the two groups *trans* to L being exchanged, it is again most likely that an S_E2(cyclic) type of transition state is involved [equation (3)]. An alternative pathway, oxidative addition followed by reductive elimination, could proceed through any one of six intermediates, (A)—(F), two of which, (A) and (B), represent extremes of the transition state of equation (3). Intermediates (C) and (D) are related to (A) and (B) by reversing the sites of the added radicals; (E) and (F) are the results of *trans* oxidative additions, unlikely under these conditions. Similarly, *cis* reductive eliminations are assumed in all cases.[26] Whilst these redox sequences are less probable than S_E2(cyclic) pathways, they cannot rigorously be excluded at present.

Ethynyl Complexes.—The reaction between *cis*-[Pt(C$\equiv$CPh)$_2$(CO)L] (L = PMePh$_2$) and *cis*-[PtCl$_2$(CO)L] in CDCl$_3$ was followed by ^{31}P n.m.r. spectroscopy at −60 °C. The bis(phenylethynyl)platinum complex was prepared *in situ* from the reaction of *cis*-[PtCl$_2$(CO)L], Hg(C$\equiv$CPh)$_2$, and [NEt$_4$]Cl,[20] and a solution of *cis*-[PtCl$_2$(CO)L] added. After several hours at −60 °C, reaction to produce a single species, [PtCl(C$\equiv$CPh)(CO)L] with Cl *trans* to L, was complete [equation (4)].



(4)

Although the reaction appears simple, the site of exchange must differ on each platinum; the ethynyl *trans* to L has exchanged with the chloride *trans* to CO. This therefore represents a major variation from the usual stereochemistry of these reactions. The low temperature at which the reaction was observed makes it very unlikely that the product, or part of it, could have resulted from rearrangement of another species. The complex [PtCl(C$\equiv$CPh)(CO)L] with L *trans* to C$\equiv$CPh has been observed under these conditions.[20]

The mixed bis(ethynyl)platinum complexes previously prepared by us [20] appeared ideal to examine further this reaction, as each co-ordination site was identifiable. Accordingly, a CDCl$_3$ solution of [Pt(C$\equiv$CMe)(C$\equiv$CPh)(CO)L] (with C$\equiv$CMe *trans* to CO) was prepared from [PtCl(C$\equiv$CMe)(CO)L] (L *trans* to Cl), Hg(C$\equiv$CPh)$_2$, and [NEt$_4$]Cl,[20] and its identity and purity verified by ^{31}P and ^{1}H n.m.r. spectroscopy. To this was added one mol equiv. of *cis*-[PtCl$_2$(^{13}CO)L′] (L′ = PMe$_2$Ph) at −60 °C, and the reaction was followed by ^{31}P n.m.r. spectroscopy. Reaction was very slow at this temperature, but after briefly warming to 0 °C, 75% of the starting materials was consumed, and three major products were apparent [equation (5)].* Thereafter other minor species became apparent, presumably from competing side reactions involving the initial products as well as the reactants. These included [PtCl(C$\equiv$CPh)(CO)L] (Cl *trans* to L), [PtCl(C$\equiv$CMe)(CO)L′] (Cl *trans* to L′), and a species with a ^{31}P n.m.r. signal at δ + 2.8 p.p.m. (J_{PPt} unresolved) which could be

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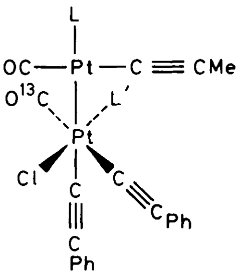
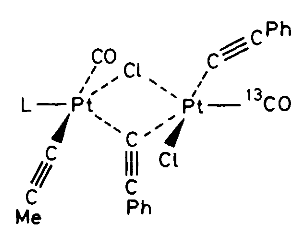
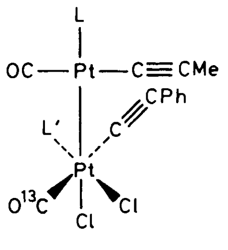
* A preliminary account of this work includes an incorrect geometry assignment of [Pt(C$\equiv$CMe)(C$\equiv$CPh)(CO)L],[27] which led to the suggestion that a specific oxidative addition/reductive elimination sequence operated for this reaction. The correct geometry [20] shown here precludes this mechanism.



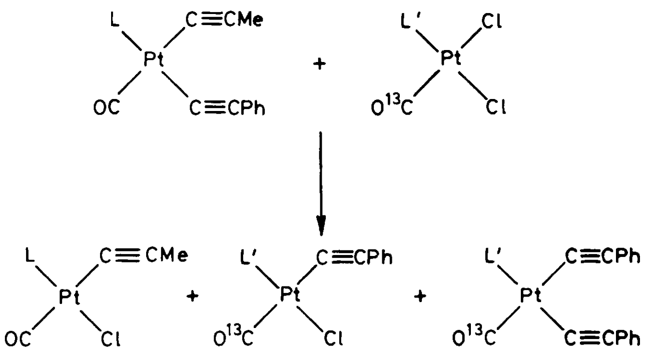
(G)



(H)



(I)



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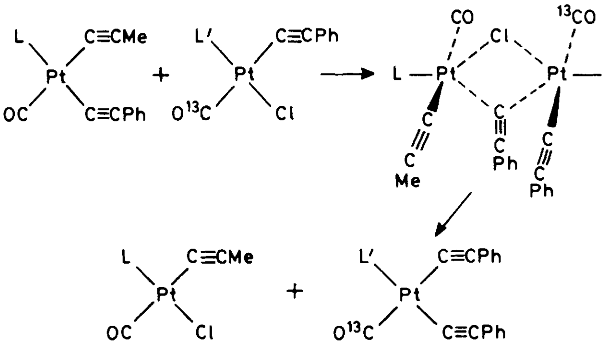

(6)

from a species with L or L′ *trans* to C≡CR.[20] The signals from these three latter species were not strong enough to determine the presence or absence of any ^{13}CO (the samples used contained 60% ^{13}C). It is clear from the major species that the carbonyls and phosphines retain their positions throughout the period of observation, so it is likely that this will apply to the secondary products also. In related examples of cyclopentadienyl–chloride exchange, carbonyl and/or phosphine scrambling are slower than the organic group and halide exchange process.[22]

The main products of equation (5), [PtCl(C≡CMe)(CO)L], [PtCl(C≡CPh)(^{13}CO)L′], and *cis*-[Pt(C≡CPh)$_2$(^{13}CO)L′], are approximately in the ratio 2 : 1 : 1, respectively. This suggests that the first two products correspond to the product of equation (4), and that the bis(phenylethynyl) material originates from the reaction between one of these products, [PtCl(C≡CPh)(^{13}CO)L′], and the starting material [Pt(C≡CMe)(C≡CPh)(CO)L]. Neither of these materials itself disproportionates to bis(phenylethynyl) complexes under the reaction conditions.[20] An interaction between [Pt(C≡CMe)(C≡CPh)(CO)L] and the other primary reaction product, [PtCl(C≡CMe)(CO)L], would produce no observable change.

The formation of *cis*-[Pt(C≡CPh)$_2$(^{13}CO)L′] can readily be accounted for by the S_E2(cyclic) mechanism [equation (6)] (or one of the related oxidative addition/reductive elimination pathways),[19,20] since the groups exchanged are both *trans* to tertiary phosphine.

Oxidative addition of [Pt(C≡CMe)(C≡CPh)(CO)L] to [PtCl(C≡CPh)(^{13}CO)L′] to produce intermediate (G) could also lead to the observed products by reductive elimination, but this is highly unlikely or an analogous addition to [PtCl(C≡CMe)(CO)L] would also be expected, leading to products different to those observed.

Formation of [PtCl(C≡CMe)(CO)L] and [PtCl(C≡CPh)(^{13}CO)L′] in equation (5) cannot so easily be rationalised. The expected S_E2(cyclic) transition state, (H), must involve migration of a group *trans* to CO. Oxidative addition of [Pt(C≡CMe)(C≡CPh)(CO)L] to *cis*-[PtCl$_2$(^{13}CO)L′] could lead to the observed products *via* intermediate (I), but there is no precedent for such a process.

We have no explanation to account for this anomaly. Since ethynyls appear to be atypical in that the isomer of [PtCl(C≡CR)(CO)L] with C≡CR *trans* to L, the usual one with other organic groups,[22] is not readily obtained,[20] it is tempting to suggest that this isomer is in fact produced in reactions (4) and (5), but rapidly converts to the isomer with Cl *trans* to L before detection. This appears unlikely, however, as we believe we have detected [PtCl(C≡CR)(CO)L] (C≡CR *trans* to L) in other reactions and under the same conditions; [20] such a rapid isomerisation is not observed.

The reaction of the other *cis*-like isomer of [Pt(C≡CMe)(C≡CPh)(CO)L] (C≡CMe *trans* to L) and *cis*-[PtCl$_2$(CO)L′] (L′ = PMe$_2$Ph) was also performed, and conforms to the same pattern. The first products detected, [PtCl(C≡CPh)(CO)L] and [PtCl(C≡CMe)(CO)L′] (both with Cl *trans* to phosphine), formed readily by brief warming to room temperature and observed at −60 °C by ^{31}P n.m.r. spectroscopy, remained the major products throughout. Further warming to room temperature (whilst some starting materials still remained) produced some *cis*-[Pt(C≡CMe)$_2$(CO)L′], as well as some [PtCl(C≡CMe)(CO)L] (Cl *trans* to L).

Experimental

Phosphorus-31 n.m.r. spectra were recorded on a Varian XL 100 spectrometer operating in the Fourier-transform mode. Hydrogen-1 n.m.r. spectra were recorded on a Perkin-Elmer R32 90-MHz spectrometer. Preparations of most of the ethynyl platinum complexes have been described previously.[20]

cis-[Pt(C≡CPh)$_2$(CO)(PMe$_2$Ph)].—*cis*-[PtCl$_2$(CO)(PMe$_2$Ph)] (17.8 mg, 0.0405 mmol), Hg(C≡CPh)$_2$ (16.3 mg, 0.0405 mmol), and [NEt$_4$]Cl (6.7 mg, 0.0405 mmol) were mixed in CDCl$_3$ (0.5 cm^3) according to published procedures.[20] Phosphorus-31 n.m.r. spectrometry revealed the formation of a single phosphine-containing *product*, *cis*-[Pt(C≡CPh)$_2$(CO)(PMe$_2$Ph)] [δ(P) −15.4 p.p.m., $^1J_{\mathrm{PPt}}$ 2 054 Hz].

cis-[Pt(C≡CMe)$_2$(CO)(PMe$_2$Ph)].—*cis*-[PtCl$_2$(CO)(PMe$_2$Ph)] (17.5 mg, 0.0405 mmol), Hg(C≡CMe)$_2$ (11.3 mg, 0.0405 mmol), and [NEt$_4$]Cl (6.7 mg, 0.0405 mmol) were similarly mixed in CDCl$_3$ (0.5 cm^3), and the single *product* observed by ^{31}P n.m.r. spectroscopy [δ(P) −15.4 p.p.m., $^1J_{\mathrm{PPt}}$ 2 063 Hz].

[PtCl(C≡CPh)(CO)(PMe$_2$Ph)] (CO trans *to* C≡CPh).—This complex was prepared by a procedure identical to that above [20] from *cis*-[PtCl$_2$(CO)(PMe$_2$Ph)] (17.4 mg, 0.0405 mmol), Hg(C≡CPh)$_2$ (8 mg, 0.020 24 mmol), and [NEt$_4$]Cl (3.3 mg, 0.020 24 mmol) in CDCl$_3$ (0.5 cm^3); ^{31}P n.m.r. parameters were δ(P) −14.4 p.p.m., J_{PPt} 3 097 Hz.

Similarly prepared was [PtCl(C≡CMe)(CO)(PMe$_2$Ph)] (CO *trans* to C≡CMe): δ(P) −15.3 p.p.m., J_{PPt} 3 108 Hz.

cis-[PtPh$_2$(CO)(PMePh$_2$)].—*cis*-[PtCl$_2$(C$_2$H$_4$)(PMePh$_2$)] (20 mg, 0.0405 mmol) and HgPh$_2$ (14.4 mg, 0.0405 mmol) were mixed in CDCl$_3$ (0.5 cm^3). A white precipitate of HgPhCl was removed by filtration from the yellow solution, shown by ^{31}P n.m.r. spectrometry to contain [Pt$_2$(μ-Cl)$_2$Ph$_2$(PMePh$_2$)$_2$] as as the only phosphine-containing compound [δ(P) −1.04 p.p.m., J_{PPt} 5 001 Hz]. Passage of CO through this solution (5 min) followed by standing at ambient temperature (15 min) resulted in a solution containing only [PtClPh(CO)(PMePh$_2$)] (CO *trans* to Ph).[23]

To this solution was added HgPh$_2$ (14.4 mg, 0.0405 mmol), and HgPhCl filtered off after 10 min. Phosphorus-31 n.m.r. spectroscopy then revealed the presence of *cis*-[PtPh$_2$(CO)(PMePh$_2$)] as the only phosphine-containing compound (δ +2.4 p.p.m., J_{PPt} 1 606 Hz).[25]

Reactions of [PtPh$_2$(CO)(PMePh$_2$)].—(*a*) *With* HgCl$_2$. A sample of *cis*-[PtPh$_2$(CO)(PMePh$_2$)] prepared as above was treated with HgCl$_2$ (one mol equiv.). A ^{31}P n.m.r. spectrum recorded immediately at ambient temperature revealed the presence only of the isomer of [PtClPh(CO)(PMePh$_2$)] with CO *trans* to Ph.[23]

(*b*) *With cis*-[PtCl$_2$(^{13}CO)(PMePh$_2$)]. To a sample of *cis*-[PtPh$_2$(CO)(PMePh$_2$)] prepared *in situ* as above was added one mol equiv. of *cis*-[PtCl$_2$(^{13}CO)(PMePh$_2$)] at room temperature. Phosphorus-31 n.m.r. examination after several minutes showed that all the starting materials had been replaced by an equimolar mixture of [PtClPh(CO)(PMePh$_2$)] (CO *trans* to Ph) and [PtClPh(^{13}CO)(PMePh$_2$)] (CO *trans* to Cl), this latter compound existing in equilibrium with [Pt$_2$(^{13}COPh)$_2$(μ-Cl)$_2$(PMePh$_2$)$_2$].[23]

[Pt(C≡CMe)(C≡CPh)(CO)(PMePh$_2$)] (C≡CMe trans *to* CO) *and cis*-[PtCl$_2$(^{13}CO)(PMe$_2$Ph)].—A solution of [Pt(C≡CMe)(C≡CPh)(CO)(PMePh$_2$)] (C≡CMe *trans* to CO) was prepared by published procedure [20] (0.0405 mmol in 0.5 cm^3 of CDCl$_3$), and its identity and purity confirmed by ^{31}P and ^{1}H n.m.r. spectroscopy. Solid *cis*-[PtCl$_2$(^{13}CO)(PMe$_2$Ph)] (17.5 mg, 0.0405 mmol) was added at −60 °C. The reaction mixture was held at −60 °C, and little change in its ^{31}P n.m.r. spectrum was noticed after 1 h. The n.m.r. tube was allowed to warm to *ca.* 0 °C for a few seconds, then cooled to −60 °C again. This resulted in 75% of the starting materials being replaced by [PtCl(C≡CMe)(CO)(PMePh$_2$)], [PtCl(C≡CPh)(^{13}CO)(PMe$_2$Ph)] (both with ethynyl *trans* to CO), and *cis*-[Pt(C≡CPh)$_2$(^{13}CO)(PMe$_2$Ph)] in ratio 2 : 1 : 1. After longer periods of warming to 0 °C, these remained the major products, but other species were also apparent from ^{31}P n.m.r. investigation. These included [PtCl(C≡CPh)(CO)(PMePh$_2$)] and [PtCl(C≡CMe)(CO)(PMe$_2$Ph)] (both with ethynyl *trans* to CO), and a species with δ(P) 2.8 p.p.m. (J_{PPt} unresolved). No ^{13}C–^{31}P coupling could be resolved in these latter species.

In an experiment similar to above, the reaction between *cis*-[Pt(C≡CPh)$_2$(CO)(PMePh$_2$)] and *cis*-[PtCl$_2$(CO)(PMePh$_2$)] was followed by ^{31}P n.m.r. spectroscopy. The reaction proceeded to completion in 2 h at 0 °C, the only product being [PtCl(C≡CPh)(CO)(PMePh$_2$)] (Cl *trans* to PMePh$_2$).

[Pt(C≡CMe)(C≡CPh)(CO)(PMePh$_2$)] (C≡CMe trans *to phosphine) and cis*-[PtCl$_2$(CO)(PMe$_2$Ph)].—*cis*-[PtCl$_2$(CO)(PMe$_2$Ph)] (15 mg, 0.0346 mmol) was added to a solution of [Pt(C≡CMe)(C≡CPh)(CO)(PMePh$_2$)] {C≡CMe *trans* to PMePh$_2$; prepared from *cis*-[PtCl$_2$(CO)(PMePh$_2$)] (25 mg, 0.0505 mmol), Hg(C≡CPh)$_2$ (10 mg, 0.0252 mmol), Hg(C≡CMe)$_2$ (7 mg, 0.0252 mmol), and [NEt$_4$]Cl (8 mg, 0.0505 mmol) in CDCl$_3$ (0.5 cm^3), and its stereochemistry confirmed by ^{31}P and ^{13}C n.m.r. spectroscopy}.[20] After mixing at room temperature, ^{31}P n.m.r. spectroscopy at −60 °C showed the presence of [PtCl(C≡CPh)(CO)(PMePh$_2$)] and [PtCl(C≡CMe)(CO)(PMe$_2$Ph)] (both with ethynyl *trans* to CO), as well as unreacted starting materials. Further brief warming to room temperature (*ca.* 1 min) resulted in almost complete consumption of the starting materials. As well as the major products already observed, *cis*-[Pt(C≡CMe)$_2$(CO)(PMe$_2$Ph)] and [PtCl(C≡CMe)(CO)(PMePh$_2$)] (Cl *trans* to PMePh$_2$) were also apparent by ^{31}P n.m.r. investigation.

References

1 R. J. Cross and R. Wardle, *J. Chem. Soc. A*, 1970, 840.
2 F. Glockling and R. J. I. Pollock, *J. Chem. Soc., Dalton Trans.*, 1974, 2259.
3 J.-E. Bäckvall and B. Åkermark, *J. Chem. Soc., Chem. Commun.*, 1975, 82.
4 A. F. Clemmit and F. Glockling, *J. Chem. Soc. A*, 1971, 1164.
5 A. Segnitz, E. Kelly, S. H. Taylor, and P. M. Maitlis, *J. Organomet. Chem.*, 1977, **124**, 113.
6 O. Rossell, J. Sales, and M. Seco, *J. Organomet. Chem.*, 1981, **205**, 133.
7 C. Eaborn, K. J. Odell, and A. Pidcock, *J. Organomet. Chem.*, 1978, **146**, 17.
8 Z. Dawoodi, C. Eaborn, and A. Pidcock, *J. Organomet. Chem.*, 1979, **170**, 95.
9 C. Eaborn, K. Kundu, and A. Pidcock, *J. Chem. Soc., Dalton Trans.*, 1981, 933.
10 C. Eaborn, K. Kundu, and A. Pidcock, *J. Chem. Soc., Dalton Trans.*, 1981, 1223.
11 J. K. Jawad, R. J. Puddephatt, and M. A. Stalteri, *Inorg. Chem.*, 1982, **21**, 332.
12 C. Eaborn, K. J. Odell, and A. Pidcock, *J. Chem. Soc., Dalton Trans.*, 1978, 357.
13 C. Eaborn, K. J. Odell, and A. Pidcock, *J. Chem. Soc., Dalton Trans.*, 1979, 758.
14 O. A. Reutov, *Tetrahedron*, 1978, **34**, 2827; M. H. Abraham, in ' Comprehensive Chemical Kinetics,' eds. C. H. Bamford and C. F. H. Tipper, Elsevier, Amsterdam, 1973, vol. 12.
15 J. Kuyper, *Inorg. Chem.*, 1977, **16**, 2171; 1978, **17**, 77.
16 J. K. Jawad and R. J. Puddephatt, *Inorg. Chim. Acta*, 1978, **31**, L391.
17 J. P. Collman and W. R. Roper, *Adv. Organomet. Chem.*, 1968, **7**, 53; J. Halpern, *Acc. Chem. Res.*, 1970, **3**, 386; T. G. Appleton, H. C. Clark, and L. E. Manzer, *J. Organomet. Chem.*, 1974, **65**, 275.
18 J. P. Collman and J. W. Kang, *J. Am. Chem. Soc.*, 1967, **89**, 844; J. P. Collman and C. T. Sears, *Inorg. Chem.*, 1968, **7**, 27.
19 R. J. Puddephatt and P. J. Thompson, *J. Chem. Soc., Dalton Trans.*, 1977, 1219.
20 R. J. Cross and J. Gemmill, preceding paper.

21 R. J. Puddephatt and P. J. Thomson, *J. Chem. Soc., Dalton Trans.*, 1975, 1810.
22 R. J. Cross and A. J. McLennan, *J. Chem. Soc., Dalton Trans.*, 1983, 359.
23 G. K. Anderson and R. J. Cross, *J. Chem. Soc., Dalton Trans.*, 1979, 1246.
24 T. G. Appleton, H. C. Clark, and L. E. Manzer, *Coord. Chem. Rev.*, 1973, **10**, 335; F. R. Hartley, *Chem. Soc. Rev.*, 1973, **2**, 163.
25 G. K. Anderson, H. C. Clark, and J. A. Davies, *Inorg. Chem.*, 1981, **20**, 3607.
26 P. S. Braterman and R. J. Cross, *Chem. Soc. Rev.*, 1973, **2**, 271.
27 R. J. Cross and J. Gemmill, *J. Chem. Soc., Chem. Commun.*, 1982, 1343; 1983, 756.

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