**Table I.** Rates of Cycloaddition of 3,4-Dimethylenefuran (**1**) and 3,4-Dimethylenethiophene (**2**) with Alkenes at 260–263 K[a]

| | **1** | | **2** | | |
|---|---|---|---|---|---|
| alkene | $10^6k$[b] | $k_{\mathrm{rel}}$ (direct)[g] | $10^6k$[b] | $k_{\mathrm{rel}}$ (direct)[g] | $k_{\mathrm{rel}}$ (compet)[h] |
| maleic anhydride[c] | 1200 | 20 | 570 | 57 | 90 |
| fumaronitrile[d] | 320 | 5.1 | 11 | 1.1 | 1.5 |
| dimethyl fumarate[e] | 63 | 1.0 | 10 | 1.0 | 1.0 |
| acrylonitrile[f] | 0.42 | 0.0067 | 0.024 | 0.0024 | 0.0083 |

[a] In $CH_3CN$ solvent. [b] In $M^{-1}\ s^{-1}$. [c] Concentration range 0–0.011 M. [d] 0–0.014 M. [e] 0–0.048 M. [f] 0–2 M. [g] Determined from absolute rate measurements (present work). [h] Determined from competition experiments (ref 6a).

The disappearance of both species could be fitted to second-order kinetics with rate constants ($2k_t$ in $M^{-1}\ s^{-1}$) for dimerization[3] as follows: for **1** ($CH_3CN$), $3.3 \times 10^{10}$; for **1** (toluene), $1.6 \times 10^{10}$; for **2** ($CH_3CN$), $9.3 \times 10^9$; for **2** (toluene), $5.5 \times 10^9$, each value being the average of four separate determinations. The probable uncertainty in the dimerization rate constants is about 50%, primarily because the extinction coefficients (in $M^{-1}\ cm^{-1}$) of the biradicals (**1**, 5300; **2**, 5400) are known to an accuracy of only about 20%.[11] The rates, especially those for the furan **1**, approach the diffusive encounter-controlled limit of about $10^{10}\ M^{-1}\ s^{-1}$ under these conditions.

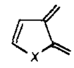
In the presence of large excesses of alkenes, the transients are quenched with pseudo-first-order kinetics. The rate constants in all cases are linear ($r \geq 0.99$ in most cases) in the alkene concentration, and the limiting rate constant at zero quencher concentration, extrapolated by a least-squares fit and converted to second-order form, is in good agreement with the dimerization rate constant determined as described above. The bimolecular rate constants are shown in Table I.

The values for the dimerization rate constants may be pertinent to the question[12] of the statistical spin effect in monoradical–monoradical reactions. Absolute rates for most of such processes seem to cluster around $2 \times 10^9\ M^{-1}\ s^{-1}$ rather than the calculated encounter-controlled limit of $10^{10}\ M^{-1}\ s^{-1}$. This has prompted the conjecture that reaction may occur in only a quarter of the encounters, since only this fraction of the radical pairs will be singlet. However, the present reaction partners are *singlet biradicals*, whose pairs necessarily are singlet. Hence product formation is spin-allowed from all encounters. Perhaps the consequent increase in efficiency of combination contributes to the exceptionally high rates observed here.

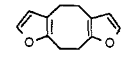
Table I shows that the effect of structure on the relative reactivities of alkenes toward the biradical **2** is essentially the same whether measured by competition experiments[6a] or by the present absolute rates (compare the last two columns). This leaves little doubt that the species responsible for the formation of cycloadducts in preparative experiments[3,6a] are indeed the purple substances[3–5,6b] whose kinetic behavior now has been observed directly.

The absolute rates of the cycloaddition reactions also are extremely fast (Table I). With stereochemically labeled alkenes, these reactions are ≥99% stereospecifically syn additions[3,11] which most probably pass over four-center, Diels–Alder-like transition states. However, the cycloaddition rates of **1** and **2** with alkenes exceed those[13] of even the most reactive Diels–Alder pairs by many orders of magnitude. For example, the **1**–maleic anhydride reaction is faster than that of cyclopentadiene–tetracyanoethylene, one of the fastest Diels–Alder cases, by a factor of about $3 \times 10^5$; the factor becomes $10^{10}$ in a structurally more closely related case, cyclopentadiene–maleic anhydride. These large enhancements of reactivity probably are caused by the small frontier orbital gap[13] in the reaction of alkenes with these electron-rich biradicals and by the much greater exothermicity (about 80 kcal/mol as compared to the 40 kcal/mol of the Diels–Alder reaction).

The dimerization rates of the species **1** and **2** greatly exceed those of their isomers, 2,3-dimethylenefuran (**8**),[14a] and 2,3-dimethylenethiophene (**9**),[14b] respectively. Compound **8**, for example, is persistent in solution at −78 °C in a concentration of about 0.2 M, from which a rough value for its dimerization rate constant of about $10^{-3}\ M^{-1}\ s^{-1}$ can be calculated. Thus, the dimerization of **1** is some $10^{13}$ times as fast as that of **8**! Although



8: X = O
9: X = S



10

the dimerization of **8**, helped by the creation of two aromatic furan units in the dimer **10**, has a substantial thermodynamic driving force (about 60 kcal/mol[15]), the exothermicity of the dimerization of the biradical **1** (about 100 kcal/mol[15]) remains much greater. One can hardly refrain from the conclusions that this difference is a major cause of the large rate effects and that the latter constitute dramatic experimental demonstrations of the consequences of non-Kekulé character.

**Acknowledgment.** This work was supported in part by the National Science Foundation.

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(12) Ingold, K. U. In *Free Radicals*; Kochi, J. K., Ed.; Wiley: New York, 1973; Vol. I, p 40 and references cited therein.

(13) For a review, see: Sauer, J.; Sustmann, R. *Angew. Chem., Intl. Ed. Engl.* **1980**, *19*, 779.

(14) (a) Trahanovsky, W. S.; Cassady, T. J.; Woods, T. L. *J. Am. Chem. Soc.* **1981**, *103*, 6691. (b) Chauhan, P. M. S.; Jenkins, G.; Walker, S. M.; Storr, R. C. *Tetrahedron Lett.* **1988**, *29*, 117.

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## Total Synthesis of Amphimedine

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*Received March 3, 1988*

The unusual structure of amphimedine (**1**), a cytotoxic alkaloid recently isolated from a Pacific sponge,[1] was elucidated by using long-range C–H and C–C NMR correlations. Amphimedine belongs to a small group of highly unsaturated fused pentacyclic alkaloids isolated from marine organisms[2] and is loosely related to the mimosamycin-type family of antibiotics.[3] In this communication we report the first total synthesis of amphimedine.

In the retrosynthetic analysis (Scheme I), amphimedine can be derived from diazaanthraquinone **2**, available by a hetero-Diels–Alder reaction[4] of 2-azadiene **3**[5,6] with the quinoline-5,8-

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(1) Schmitz, F. J.; Agarwal, S. K.; Gunasekera, S. P.; Schmidt, P. G.; Shoolery, J. N. *J. Am. Chem. Soc.* **1983**, *105*, 4835.

(2) (a) 2-Bromoleptoclinidinone: Bloor, S. J.; Schmitz, F. J. *J. Am. Chem. Soc.* **1987**, *109*, 6137. (b) Calliactine: Cimino, G.; De Rosa, S.; De Stefano, S.; Sodano, G. *Pure Appl. Chem.* **1986**, *58*, 375.

(3) For a review, see: Arai, T.; Kubo, A. *The Alkaloids*; Brossi, A., Ed.; Academic Press: New York, 1983; Vol. 21, Chapter 3.

(4) For recent reviews, see: (a) Kametani, T.; Hibino, S. *Adv. Heterocycl. Chem.* **1987**, *42*, 245. (b) Boger, D. L. *Tetrahedron* **1983**, *39*, 2869. (c) Weinreb, S. M.; Staib, R. R. *Tetrahedron* **1982**, *38*, 3087.

(5) Sainte, F.; Serckx-Poncin, B.; Hesbain-Frisque, A.-M.; Ghosez, L. *J. Am. Chem. Soc.* **1982**, *104*, 1428.

(6) For a synthesis of the antibiotic mimosamycin based on a cycloaddition reaction with **3**, see: McKillop, A.; Brown, S. P. *Synth. Commun.* **1987**, *17*, 657.

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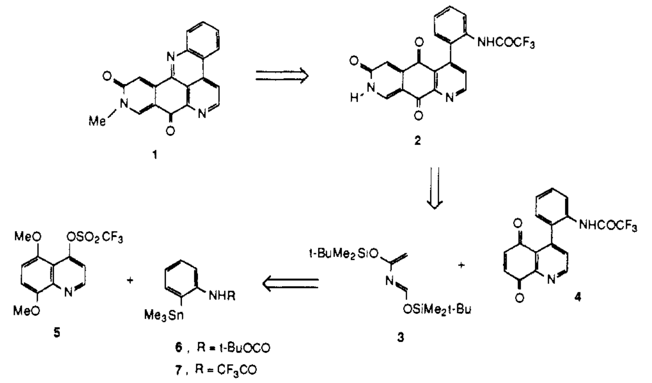
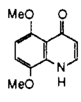
**Scheme I**

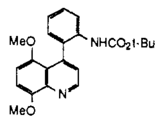


dione **4**.[7] The desired regiochemistry was expected to arise by attack of the more nucleophilic end of the electron-rich diene (C-4) at the C-6 position of the dienophile.[7a,8] The synthesis of **4** takes advantage of our recently developed palladium-catalyzed cross-coupling reaction of aryl triflates with organostannanes.[9]

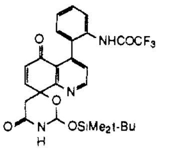
Reaction of the readily available quinolinone **8**[10] with trifluoromethanesulfonic anhydride (1.2 equiv) in dichloromethane containing 2,6-lutidine (1.4 equiv) and *N*,*N*-dimethylaminopyridine (0.2 equiv) at 0 °C for 2 h, at 0 to 23 °C for 30 min, and at 23 °C for 1 h furnished the desired triflate **5** in 92–95% yield. Palladium-catalyzed reaction[9] of **5** with organostannane **6**[11] (1.3 equiv) in 1,4-dioxane at 100 °C in the presence of $Pd(PPh_3)_4$ (2 mol %) and LiCl (3 equiv) for 5–7 h gave 4-arylquinoline **9** in 87% yield. Similarly, reaction of **5** with stannane **7**[12,13] proceeded somewhat more sluggishly (100 °C, 16 h) to afford **10** in 71% yield. Quinoline **9** was readily converted into **10** in two steps: (i) cleavage of the carbamate protecting group in trifluoroacetic acid at 23 °C for 1 h to yield aniline **11** (94–100%), and (ii) acylation with trifluoroacetic anhydride (1.0 equiv) and diisopropylethylamine (1.0 equiv) in tetrahydrofuran at 0 °C for 30 min (100%).



8



9, R = t-BuOCO
10, R = CF3CO
11, R = H



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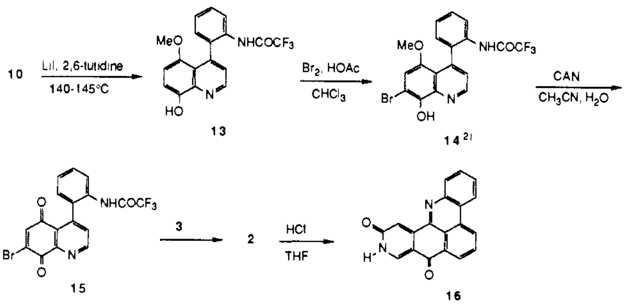
Oxidation of **9** with ceric ammonium nitrate (CAN)[14] (2–3 equiv) or silver(II) oxide[15] failed to provide any of the expected quinone as a consequence of the competitive oxidation of the 4-aryl ring. However, exposure of **10** to CAN (2.4 equiv) in 2:1 acetonitrile–water at 23 °C for 15 min yielded quinoline-5,8-dione **4** in 85% yield.[16]

Reaction of **4** with azadiene **3**[5] (2.5 equiv) in dry and acid-free $CHCl_3$ at 23 °C for 6 h followed by selective desilyation with methanol (23 °C, 30 min) yielded cycloadduct **12**[17] as a single product in 64% yield. The structure of **12** was assigned on the basis of the spectroscopic data[18] and on the assumption that the attack of **4** had taken place on the less hindered and more electron-defficient C-8 carbonyl group.[7a] However, when the cycloaddition was carried out in tetrahydrofuran (THF) solution (23 °C, 6 h) the desired diazaanthraquinone **2** was isolated in 48% yield[19] after treatment of the crude reaction mixture with pyridinium hydrofluoride (5 equiv). The regiochemistry of **2** was assigned on the basis of literature precedent[7a] and was further correlated by the unambiguous synthesis shown below.

Selective demethylation of **10** afforded **13**[20] in 64% yield. Bromination of **13** followed by CAN (2.0 equiv) oxidation at 0 °C for 15 min gave rise to **15**[21] in 59% yield (two steps). Hetero-Diels–Alder reaction of **15** with **3** in THF at 23 °C for 21 h afforded **2**[22] (47%), identical in all respects with a sample obtained from **4**.



The synthesis was completed by acid hydrolysis of **2** (1:1 6 M HCl–THF, 70–80 °C, 3 h) to yield desmethylamphimedine **16** in 86% yield as a yellow solid. Reaction of **16** with $Me_2SO_4$ (6

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(7) For the Diels–Alder reaction of quinolinequinones, see: (a) Potts, K. T.; Bhattacharjee, D.; Walsh, E. B. *J. Org. Chem.* **1986**, *51*, 2011; *J. Chem. Soc., Chem. Commun.* **1984**, 114. (b) Link, H.; Bernauer, K.; Englert, G. *Helv. Chim. Acta* **1982**, *65*, 2645.

(8) A moderate preference for C-6 attack has been reported in the reaction of anilines with quinoline-5,8-dione. Addition of an acid catalyst (HOAc or $CeCl_3 \cdot 7H_2O$) promoted almost exclusive C-6 attack: Pratt, Y. P. *J. Org. Chem.* **1962**, *27*, 3905.

(9) Echavarren, A. M.; Stille, J. K. *J. Am. Chem. Soc.* **1987**, *109*, 5478.

(10) Synthesized in two steps from 2,5-dimethoxyaniline (71% yield) by a published procedure: Cassis, R.; Tapia, R.; Valderrama, J. A. *Synth. Commun.* **1985**, *15*, 125. The $^1H$ NMR ($Me_2SO$-$d_6$) of **8** differs with that reported by Cimino et al. in the NH chemical shift (δ 10.97 instead of the reported 3.33) and the multiplicity of the H-2 signal (brd vs t): Cimino, G.; De Rosa, S.; De Stefano, S.; Spinella, A.; Sodana, G. *Tetrahedron Lett.* **1984**, *25*, 2925.

(11) (a) Synthesized in 70–75% yield by ortho-lithiation of *N*-*tert*-butoxycarbonylaniline[10b] [t-BuLi (2.4 equiv), THF, −78° to −20°C] and reaction with chlorotrimethylstannane (1.5 equiv). (b) Muchowski, J. M.; Veruti, M. C. *J. Org. Chem.* **1980**, *45*, 4798.

(12) (a) Synthesized in ca. 20% yield from trimethylstannane **6**[11b] by the sequence: (i) Catalytic hydrogenation ($H_2$, 1 atm), 10% Pd/C, EtOH and (ii) acylation with trifluoroacetic anhydride and diisopropylethylamine in THF.[10c] (b) Bumagin, N. A.; Bumagina, I. G.; Beletskaya, I. P. *Dokl Akad. Nauk SSSR* **1984**, *274*, 1103; (Engl. Transl.) **1984**, *274*, 53. (c) Extensive destannylation takes place during this process. The coupling reaction was carried out with crude stannane **7** containing trifluoroacetanilide.

(13) Halogen-lithium exchange (Cf: Wender, P. A.; White, A. W. *Tetrahedron* **1983**, *39*, 3767) on 2-bromo or 2-iodotrifluoroacetanilide and reaction with the chlorotrimethylstannane provided **7** in poor yields (5–10%). Similar results were obtained from 2-bromonitrobenzene (Cf: Parham, W. E.; Piccirili, R. M. *J. Org. Chem.* **1976**, *41*, 1268).

(14) Jacob III, P.; Callery, P. S.; Shulzin, A. T.; Castagnoli, N., Jr. *J. Org. Chem.* **1976**, *41*, 3627.

(15) Snyder, C. D.; Bondinell, W. E.; Rapoport, H. *J. Org. Chem.* **1971**, *36*, 3955.

(16) An alternative route to **4** was carried out by the sequence: (i) acylation of **9** with trifluoroacetic anhydride and triethylamine to yield a labile imide, (ii) in situ oxidation with CAN (32% overall yield for the two steps), and (iii) cleavage of the carbamate with trifluoroacetic acid at 20 °C for 2 h (92%).

(17) This mode of reaction of a diene with a naphthoquinone is, to the best of our knowledge, unprecedented. For a photoinduced Diels-Alder reaction of benzoquinone with acyclic dienes to yield spiro adducts, see: Barltrop, J. A.; Hesp, B. *J. Chem. Soc.* **1965**, 5182.

(18) White solid; mp: 176–178 °C (4:1 EtOAc-hexanes); $^1H$ NMR ($CDCl_3$, 270 MHz) δ 8.98 (d, J = 4.6 Hz, 1 H), 7.93 (dd, J = 8.3, 1.2 Hz, 1 H), 7.72 (brs, 1 H), 7.67 (d, J = 10.5 Hz, 1 H), 7.54 (td, J = 7.8, 1.5 Hz, 1 H), 7.39 (td, J = 7.6, 1.1 Hz, 1 H), 7.33 (d, J = 4.6 Hz, 1 H), 7.24 (dd, J = 7.6, 1.6 Hz, 1 H), 6.56 (d, J = 10.5 Hz, 1 H), 6.04 (br, 1 H), 5.59 (d, J = 1.9 Hz, 1 H; collapses to a singlet on irradiation at δ 6.04), 3.13 (d, J = 17.4 Hz, 1 H), 2.58 (d, J = 17.3 Hz, 1 H), 0.87 (s, 9 H), 0.09 (s, 3 H), 0.06 (s, 3 H). $^{13}C$ NMR ($CDCl_3$, 68 MHz) δ 181.14, 164.02, 151.42, 148.73, 146.74, 146.06, 137.15, 132.59, 132.46, 130.06, 129.71, 126.79, 126.57, 123.90, 95.00, 73.78, 57.44, 25.33, 17.68, −4.74, −5.35. IR (KBr) 3240, 3060, 2960, 2860, 1740 sh, 1725, 1690, 1680, 1580, 1470, 1440, 1320, 1195, 1150, 1035, 995, 895, 830-820, 770, 745 $cm^{-1}$. Anal. Calcd for $C_{26}H_{28}F_3O_5Si$: C, 57.02; H, 5.15. Found: C, 56.96; H, 5.19.

(19) No cycloaddition was observed in the presence of added Lewis acids (i.e. $BF_3 \cdot OEt_2$, $CeCl_3$).

(20) In the $^1H$ NMR ($CDCl_3$) of **10** well separated methoxyl resonances were observed at δ 4.10 (C-8 methoxy group) and δ 3.44 (C-5 methoxy group, aryl-shielded). **14** showed a methoxyl resonance at δ 3.45.

(21) The regiochemistry of **14** was supported by the observation of a 14% enhancement of the H-6 signal in an $^1H$ NOEDIFF experiment involving irradiation of the methoxy group hydrogens.

(22) For the regiochemistry of the reaction of 2-haloquinones with activated dienes, see: Boisvert, Z.; Brassard, P. *Tetrahedron Lett.* **1983**, *24*, 2453, and references cited therein.

equiv) and $K_2CO_3$ (3 equiv) in DMF at 23 °C for 3 h gave rise to amphimedine **1** in 96% yield as a dark yellow solid of mp >300 °C, identical in all aspects with an authentic sample by $^1H$ and $^{13}C$ NMR, IR, silica gel-TLC analysis in several solvent systems, and mass spectral comparison.

In summary, a concise synthesis of the alkaloid amphimedine has been achieved in eight steps (21-23% overall yield). This synthesis illustrates the utility of the palladium-catalyzed cross-coupling reaction of organostannanes with functionalized electrophiles in the elaboration of complex molecules. During the course of this study a dramatic solvent effect in a hetero-Diels-Alder reaction has been uncovered.

**Acknowledgment.** This research was supported by a grant (CHE-8703218) from the National Science Foundation. Antonio M. Echavarren acknowledges the receipt of a NATO Fellowship. The palladium was provided under the Johnson-Matthey Metal Loan Program. We are grateful to Professor F. J. Schmitz for a sample of amphimedine and copies of the $^1H$ and $^{13}C$ NMR spectra.

**Supplementary Material Available:** Spectroscopic, physical, and analytical data for new compounds and experimental procedures for the preparation of **2**, **16**, and **1** (5 pages). Ordering information is given on any current masthead page.

## Electronic Emission from Bis(bridging diphosphine) Derivatives of $Re_2(CO)_{10}$. Characterization of the Emissive State as $^3(d\sigma \rightarrow d\sigma^*)$

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*Received May 12, 1987*

We recently[1] reported electronic emission from $d^7$-$d^7$ singly metal-metal bonded compounds of the type $Pt^{III}_2(pop)_4L_2{}^{n-}$, where pop is $P_2O_5H_2{}^{2-}$ and L is, for example, a halide. The emission was unequivocally characterized as the first observation of emission from a $d^7$-$d^7$ metal-metal excited state by observation of a long progression in $\nu(Pt_2)$ for the low-temperature emission of the $K^+$ salt of the L = Br derivative. We have subsequently observed[2] similar emission for the previously reported[3] $d^7$-$d^7$ compounds $Ir^{II}_2(TMB)_4L_2{}^{n+}$, where TMB is 2,5-diisocyano-2,5-dimethylhexane.

Common factors in these compounds are bridging ligands (presumably preventing metal-metal dissociation) and third transition series metal ions (presumably enhancing spin-orbit coupling, hence radiative transitions from a low-energy metal-metal triplet-parentage state).

The compounds $M_2(CO)_{10}$ M = Mn, Tc, Re, have been the classic examples of highly dissociative metal-metal excited states, yielding in the case of $Mn_2(CO)_{10}$, $Mn(CO)_5$ radicals on the time scale of less than 20 ps in the gas phase.[4] The spectroscopy, photophysics, and photochemistry of these dimers has been exhaustively studied.[5] It is therefore of great interest to determine whether bridging ligands might enforce a long lived emissive state by preventing the homolysis pathway. Establishing a correlation with these compounds would make it completely clear that the same type of excited states are being considered in the case of the previously reported Pt(III) dimers. Guided by our previous work, we have investigated derivatives of $Re_2(CO)_{10}$. We now report that derivatives involving substitution of 2 equiv of either[6] bis(dimethylphosphino)methane (dmpm) or bis(diphenylphosphino)methane (dppm) are highly emissive at low temperature. In contrast, under the same conditions we find that both $Re_2(CO)_{10}$ and $Re_2(CO)_8(P(C_6H_5)_3)_2$ are nonemissive, consistent with the photophysical results.[4]

**Table I.** Emission Data for $Re_2(P\text{-}P)_2(CO)_6$ Compounds at 77 K

| | | | 2-methylpentane glass | |
|---|---|---|---|---|
| P-P | λ (nm) | solid τ (μs) | λ (μm) | τ (μs) |
| dmpm | 705 | 23 | 690 | 31 |
| dppm | 620 | 64 | 610 | 76[a] |

[a] In 2-methyltetrahydrofuran glass (49 μs). The absorption and emission spectra are insensitive to solvent.

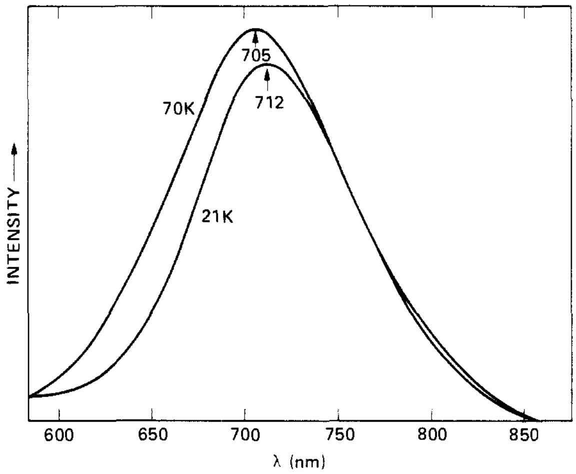

**Figure 1.** Emission spectrum of a solid sample of $Re_2(dmpm)_2(CO)_6$ at 20 and 70 K. The spectral slit width was 2 nm, and the exciting wavelength was 400 nm.

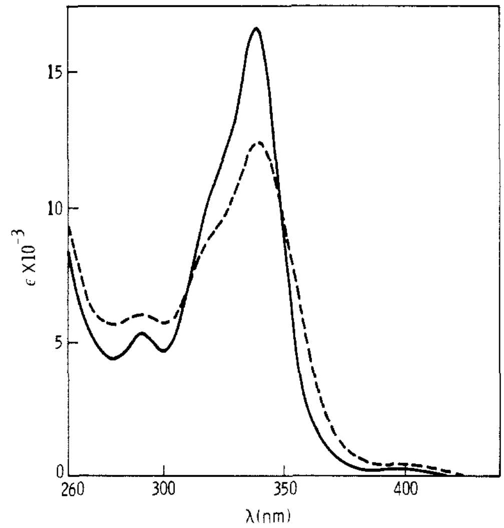

**Figure 2.** The absorption spectrum of $Re_2(dmpm)_2(CO)_6$ at room temperature (--) and 77 K (—) in 2-methylpentane solution. The 77 K spectrum has been corrected for solvent contraction.

We have concentrated upon the dmpm derivative, $Re_2(dmpm)_2(CO)_6$ (I), because the absence of phenyl groups clarifies the electronic spectrum. In Figure 1 we show the emission

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[1] Stiegman, A. E.; Miskowski, V. M.; Gray, H. B. *J. Am. Chem. Soc.* **1986**, *108*, 2781-2782.

[2] Stiegman, A. E.; Miskowski, V. M.; Gray, H. B., unpublished work.

[3] Miskowski, V. M.; Smith, T. P.; Loehr, T. M.; Gray, H. B. *J. Am. Chem. Soc.* **1985**, *107*, 7925-7934.

[4] (a) Freedman, A.; Bersohn, R. *J. Am. Chem. Soc.* **1978**, *100*, 4116-4118. (b) Rothberg, L. J.; Cooper, N. J.; Peters, K. S.; Vaida, V. *J. Am. Chem. Soc.* **1982**, *104*, 3536-3537.

[5] Two recent reviews are as follows: (a) Meyer, T. J.; Caspar, J. V. *Chem. Rev.* **1985**, *85*, 187-218. (b) Miskowski, V. M.; Gray, H. B., In *Understanding Molecular Properties*; Avery, J., Dahl, J. B., Eds.; Reidel Publishing Co.: Holland, 1987; pp 1-16.

[6] These compounds were prepared following published procedures: dmpm and (King, R. B.; Raghuveer, K. S. *Inorg. Chem.* **1984**, *23*, 2482-2491) dppm (Lee, K. W.; Brown, T. L. *Organometallics* **1985**, *4*, 1025-1030).

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