that is responsible for the edge-bicapped tetrahedral coordination of Mn(2) in the trinuclear complex. A similar interaction pattern has been detected in some lithium-coordinated transition-metal derivatives.[30] The bond indices between the terminal manganese centers and the carbon atoms in the organic frameworks are comparable with the Wiberg indices found for the open ferrocenes.[4] This is also true for the slight alternancy in the pentadienyl carbon framework.

**Concluding Remarks.** The data described herein provide some interesting contrasts and comparisons between the original compound formulation of $Mn^{2+}[Mn(3\text{-}C_6H_9)_4]^-$ and the more sophisticated description developed by the theoretical studies, particularly with respect to placement of electrons and net charges. Nevertheless, both sets of results are reasonably consistent with one another in terms of total unpaired electrons in the ground state and the weak covalent bonding interactions expected between the central manganese atom and the nearby atoms in its *edge*-bicapped tetrahedral geometry. To some extent the difference between the two models may be related to the situation for metal hydrides, where the hydrogen atom may behave more as an acid or as a hydride depending on circumstance. In either case, the same metal and hydrogen orbitals are involved, and the actual charges are dependent on the relative proportions of the metal and hydrogen orbitals contributing to the ground state orbital. Thus, the simple ionic formulation and the more sophisticated theoretical sextet model may really bear some resemblance to one another after all. Further efforts to gain a better understanding of this system are in progress.

**Acknowledgment.** The work at Heidelberg has been supported by the Fonds der Chemischen Industrie, the Stiftung Volkswagenwerk, and the BASF Aktiengesellschaft in Ludwigshafen. The work at Utah has been supported by the National Science Foundation (Grant No. CHE-8120683), the donors of the Petroleum Research Fund, administered by the American Chemical Society, and the University of Utah Research Committee. R.G. and R.D.E. are further grateful to NATO for a travel grant, without which the pursuit of these studies would have been much more difficult. We thank Prof. Tobin Marks and Dr. Tamotsu Inabe of Northwestern University for helping us obtain the magnetic measurements.

**Registry No.** $Mn_3(3\text{-}CH_3C_5H_6)_4$, 85976-94-9; $MnCl_2$, 7773-01-5; Mn, 7439-96-5; potassium 3-methylpentadienyl, 74206-00-1.

**Supplementary Material Available:** A listing of observed and calculated structure factor amplitudes and Tables II and III, showing non-hydrogen atom thermal parameters and hydrogen atom positional and thermal parameters (22 pages). Ordering information is given on any current masthead page.

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(30) Böhm, M. C.; Gleiter, R. *J. Organomet. Chem.* **1982**, *228*, 1.

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Contribution from the Department of Hydrocarbon Chemistry,
Faculty of Engineering, Kyoto University, Kyoto 606, Japan

# ESR Study of the Electronic Structure of $Co_3(\eta^5\text{-}C_5H_5)_3(\mu_3\text{-}CPh)_2$

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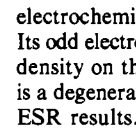
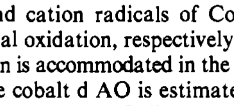
*Received April 7, 1983*

The anion and cation radicals of $Co_3(\eta^5\text{-}C_5H_5)_3(\mu_3\text{-}CPh)_2$ are generated by reduction with potassium metal and by electrochemical oxidation, respectively. The frozen-solution ESR spectra of the anion show the $Co_3$ triangle to be regular. Its odd electron is accommodated in the $a_2'$ MO ($D_{3h}$ point group) constructed mainly from cobalt d AO's, and the odd-electron density on the cobalt d AO is estimated as 0.18. The ESR spectrum of the cation suggests that the odd-electron orbital is a degenerate one. An MO diagram constructed by using the method of Dahl and co-workers is consistent with the present ESR results. The odd-electron distribution in the present anion radical is compared with those of closely related $Co_3(CO)_9(\mu_3\text{-}CPh)^-\cdot$ and $Co_3(CO)_9S\cdot$.

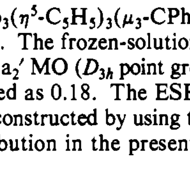
Elucidation of electronic structures and assignment of frontier orbitals of triangular $M_3$ clusters with $\mu_3$ ligand(s) are being developed by Dahl and co-workers by analyses of geometrical deformations caused by changes in the number of valence electrons[1] and by an ESR study of $Co_3(CO)_9S\cdot$[2] (**1**), which is isoelectronic to $Co_3(CO)_9(\mu_3\text{-}CPh)^-\cdot$ (**2**$^-$). The latter tricobalt complex has been examined by Robinson and his colleagues.[3] ESR studies of ion radicals of metal clusters with higher geometrical symmetries are expected to reveal electronic characteristics complementary to those obtained by studies of geometrical effects.



1



2



3

Cp = $C_5H_5$

Herein is presented our ESR study of ion radicals derived from $Co_3(\eta^5\text{-}C_5H_5)_3(\mu_3\text{-}CPh)_2$ (**3**), which has a pseudo-$D_{3h}$ geometry.[4] Our results fully support the method of constructing a qualitative molecular orbital (MO) diagram to reproduce frontier orbitals developed by Dahl and co-workers.[2,5] The d character of the odd-electron orbital of **3**$^-$ is

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(1) (a) Mag, J. J.; Ral, A. D.; Dahl, L. F. *J. Am. Chem. Soc.* **1981**, *104*, 3054 and references cited therein. (b) See also: Schilling, B. E. R.; Hoffmann, R. *Ibid.* **1979**, *101*, 3456.

(2) Strouse, C. E.; Dahl, L. F. *Discuss. Faraday Soc.* **1969**, *47*, 93; *J. Am. Chem. Soc.* **1971**, *93*, 6032.

(3) Peake, B. M.; Rieger, P. M.; Robinson, B. H.; Simpson, J. *Inorg. Chem.* **1979**, *18*, 1000.

(4) The trigonal-bipyramidal core of $Co_3(\eta^5\text{-}C_5H_5)_3(\mu_3\text{-}CSiMe_3)(\mu_3\text{-}C_3SiMe_3)$ has a pseudo-$D_{3h}$ geometry: Fritch, J. R.; Vollhardt, K. P. C.; Thompson, M. R.; Day, V. W. *J. Am. Chem. Soc.* **1979**, *101*, 2768.

(5) Vahrenkamp, H.; Uchtman, V. A.; Dahl, L. F. *J. Am. Chem. Soc.* **1968**, *90*, 3272.

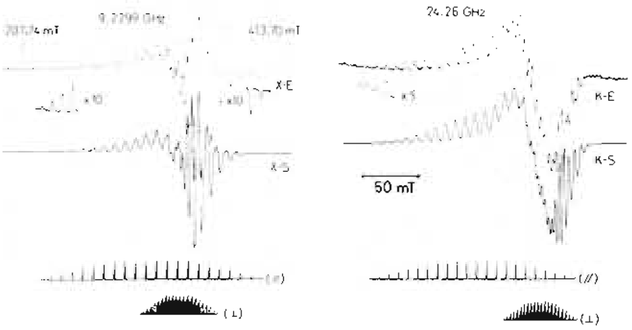


**Figure 1.** Experimental (E) and simulated (S) ESR spectra of $Co_3(\eta^5\text{-}C_5H_5)_3(\mu_3\text{-}CPh)_2^{-}\cdot$ ($3^-$) at X and K bands.

Table I. ESR Parameters of Paramagnetic Tricobalt Clusters

| radical | $g_\parallel{}^a$ | $g_\perp$ | $A_\parallel(Co)^a$/ $10^{-4}$ cm$^{-1}$ | $A_\perp(Co)$/ $10^{-4}$ cm$^{-1}$ | $\langle g\rangle^b$ | $\langle a(Co)\rangle^b$/ $10^{-4}$ cm$^{-1}$ |
|---|---|---|---|---|---|---|
| $1^c$ | 2.04 | $2.02^d$ | −69.5 | $-8.6^d$ | 2.022 | −28.9 |
| $2^{-\,e}$ | 1.996 | $2.02^d$ | −73.6 | $-13.6^d$ | 2.013 | −33.6 |
| $3^{-\,f}$ | $2.129_4$ | $1.987_5$ | −82.8 | −27.4 | 2.035 | −40.9 |
| $3^{+\,f,g}$ | 2.2 | | | | | |

[a] From solid-state spectra. [b] From solution spectra. [c] Reference 2. [d] Deduced from parallel features in solid-state spectra and isotropic parameters for solution spectra. [e] Reference 3. [f] Present work. [g] Abnormally broad; $\Delta H_{msl} = 27$ mT.

estimated and compared with those of pseudoisoelectronic **1** and **2**$^-$.

## Results

**$Co_3(\eta^5\text{-}C_5H_5)_3(\mu_3\text{-}CPh)_2^{-}\cdot$ ($3^-$).** When a 2-methyltetrahydrofuran (MTHF) solution of **3** comes in contact with potassium metal under vacuum at room temperature, a paramagnetic cobalt complex is formed. The solution at room temperature exhibits an isotropic ESR spectrum split into a 22-fold multiplet of 40.9 × $10^{-4}$ cm$^{-1}$ due to three equivalent cobalt nuclei (100% natural abundance, $I = {}^7/_2$) centered at $g = 2.035$. The spectrum is attributed to its anion radical, **3**$^-$. Anisotropic spectra of **3**$^-$ in frozen MTHF solutions at 77 K are obtained at X and K bands (Figure 1). These frozen-solution spectra fit an axially symmetric spin Hamiltonian in which three cobalt nuclei are magnetically equivalent and all the unique axes of the **g** and hyperfine splitting tensors of the three cobalt nuclei are parallel to each other. Their principal values are listed in Table I together with those for closely related **1** and **2**$^-$.[2,3]

Some comments are needed for the signs in the cobalt splitting tensor of **3**$^-$. A comparison between the isotropic splitting constant of the cobalt nucleus and the principal values of the cobalt hyperfine splitting tensor shows that the three principal values of the tensor have identical signs. The most likely choice is the negative sign (vide infra).

**$Co_3(\eta^5\text{-}C_5H_5)_3(\mu_3\text{-}CPh)_2^{+}\cdot$ ($3^+$).** The cyclic voltammogram of **3** in acetonitrile in Figure 2a displays an oxidation couple at $E_{1/2} = 0.34$ V vs. the saturated calomel electrode (SCE). The difference between the oxidation and reduction peak potentials is 70 mV, and the ratio between the peak currents is 1.0 ± 0.1. These results show that this couple is a quasi-reversible one-electron process.

When **3** in acetonitrile containing $n$-$Bu_4NClO_4$ as supporting electrolyte is oxidized at 0.5 V vs. SCE, approximately 1 faraday/mol of electricity is consumed. The cyclic voltammogram of the oxidized solution displayed in Figure 2b shows the chemical reversibility between **3** and **3**$^+$ as well as the stability of **3**$^+$. These are further supported by an almost quantitative recovery of **3** from the oxidized solution after its reduction at +0.20 V vs. SCE.

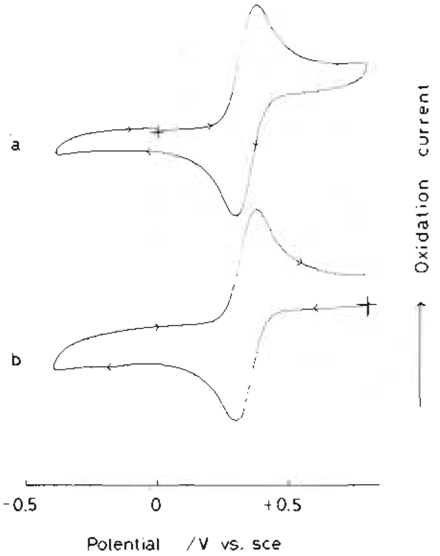


**Figure 2.** Cyclic voltammograms of **3** (a) and **3**$^+$ (b) in acetonitrile.

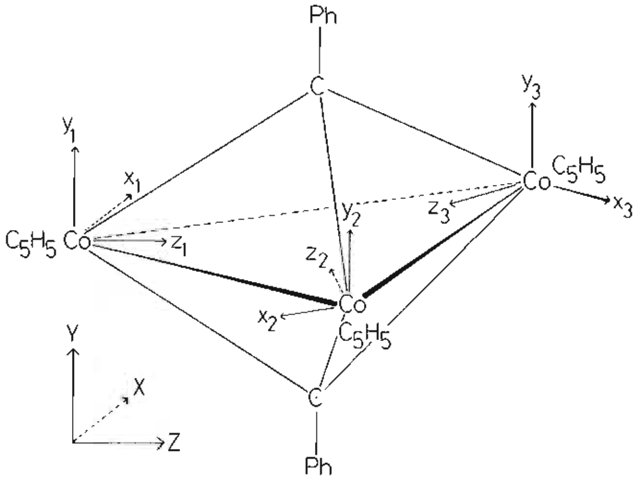


**Figure 3.** Molecular and local axes for $Co_3(\eta^5\text{-}C_5H_5)_3(\mu_3\text{-}CPh)_2$ (**3**).

The X-band ESR spectrum at 77 K of **3**$^+$, generated by electrochemical oxidation in acetonitrile, shows an abnormally broad ($\Delta H_{msl} = 27$ mT) signal centered at $g \simeq 2.2$ without any resolved hyperfine structure.

## Discussion

**The Symmetry of the Odd-Electron Orbital.** The large magnitudes of $A_\parallel(Co)$ and $A_\perp(Co)$ and the large shifts of $g_\parallel$ and $g_\perp$ from the free-electron value of $g_e = 2.0023$ indicate that the odd-electron orbital of **3**$^-$ is constructed mainly from cobalt d atomic orbitals (AO). For further analyses, we define molecular axes, $X$, $Y$, and $Z$ and local axes $x_i$, $y_i$, and $z_i$ centered on the $i$th ($i$ = 1–3) cobalt nucleus as shown in Figure 3. Cobalt AO's are designated as $p_{zi}$, $d_{yzi}$, and so on, where the former AO is the 4p AO on the $i$th cobalt atom with the axis parallel to $z_i$ and the latter AO is the 3d AO on the $i$th cobalt atom lying the $y_iz_i$ plane.

Since the observed ESR spectra of **3**$^-$ show no abnormal temperature dependence nor abnormal broadening, the odd-electron orbital is reasonably concluded to be a nondegenerate MO. The most reasonable odd-electron orbital is the MO constructed from a linear combination of $d_{xzi}$ AO's; otherwise the unique axes of the axially symmetric splitting tensors of the three cobalt nuclei cannot be parallel to each other. Because of the $D_{3h}$ geometry of **3**$^-$, the odd-electron orbital satisfying the above-mentioned restriction can only be the $a_2'$ MO of the all-antibonding combination of the three $d_{xzi}$ AO's

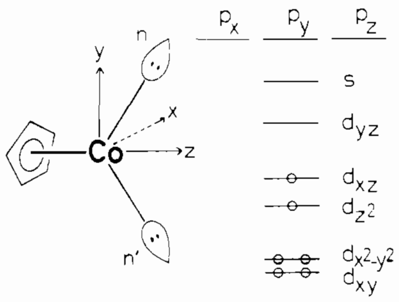
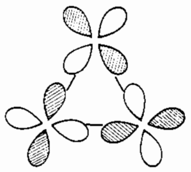


**Figure 4.** $Co^{3+}(C_5H_5)^-nn'$ fragment and its electronic configuration.

with equal weights. Thus, the ∥ axis for the spin Hamiltonian is the molar $Y$ axis, and the ⊥ axes are the $X$ and $Z$ molecular axes.



It should be pointed out that the cobalt 4s AO's are symmetry forbidden to mix into this odd-electron orbital. The isotropic splitting due to the cobalt nuclei arises from inner-shell spin polarizations, and the isotropic splitting constant should be negative.[6] All the principal values of the cobalt splitting tensor are, therefore, concluded to have negative signs.

**Qualitative MO Diagram for 3.** The method of Dahl and his colleagues[2,5] of constructing an MO diagram is applied to **3** to reproduce its frontier orbitals. The method corresponds to a development of the 18-electron rule and assumes a metal–ligand $\sigma$ bond to be formed by donation of a lone pair of electrons on the ligand to a vacant orbital on the metal atom. This assumption leads to formal oxidation numbers of +3, −3, and −1 for the cobalt atom, the carbyne group, and the cyclopentadienyl moiety, respectively. We start from a cnstruction of an MO diagram of a mononuclear fragment. $Co^{3+}(C_5H_5)^-nn'$, where n and n′ are lone pairs on the apical carbyne groups expanding to the cobalt atom (Figure 4).

Of the nine cobalt valence AO's, five AO's ($s_i$, $p_{xi}$, $p_{yi}$, $p_{zi}$, and $d_{yzi}$ are considered to be used to accept three pairs of cyclopentadienide $\pi$ electrons and lone-pair electrons of n and n′. These cobalt AO's are vacant (or dominant constituents of vacant MO's with metal–ligand $\sigma$-antibonding characters). Cobalt $d_{x^2-y^2i}$ and $d_{xyi}$ orbitals are stabilized by interaction with the vacant $\pi$ MO's of the cyclopentadienide.[7] Six d electrons of $Co^{3+}$ result in a configuration of $(d_{x^2-y^2i})^2(d_{xyi})^2(d_{z^2i})^1(d_{xzi})^1$. The present cobalt fragment is isolobal to $CH_2$.

As in the Walsh model of constructing cyclopropane from three $CH_2$ fragments,[8,1b] the MO's of **3** with dominant d characters can be constructed from orbitals of three mononuclear fragments as shown in Figure 5. On the basis of a hypothesis that metal–ligand interactions are larger than metal–metal interactions,[9] we can place $1a_2''$ and $1e''$ MO's derived from $d_{yzi}$ AO's above the MO's constructed from the other d AO's because the $d_{yzi}$ AO's are alreay used for metal–ligand bond formation. To reproduce the present ESR results of $\mathbf{3}^-$, the $1a_2'$ MO is located below the $2e'$ MO's.

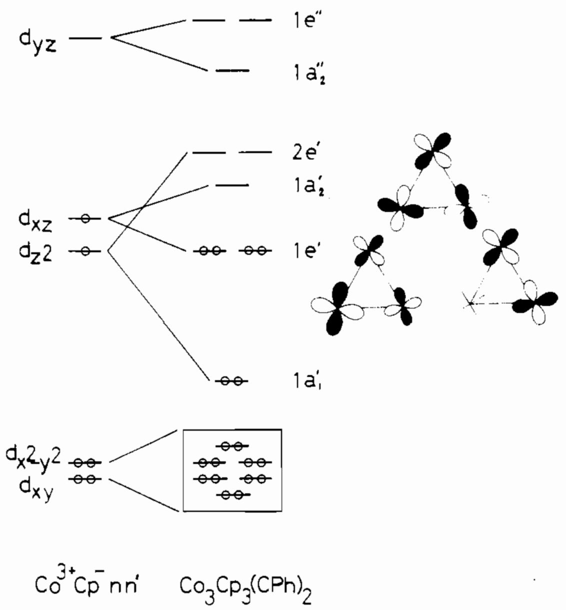


**Figure 5.** Construction of MO's for **3** from orbitals of three $Co^{3+}(C_5H_5)^-nn'$ fragments.

The diagram predicts the HOMO of **3** would be the pair of $1e'$ orbitals. The cation radical $\mathbf{3}^+$ is, therefore, predicted to have the electronic configuration of $...(1e')^3$ and to be distorted because of Jahn-Teller effects. The ESR spectrum of the cation radical, $\mathbf{3}^+$, is thus expected to be an abnormally broad one with a $g$ factor with a large positive shift from $g_e$. This expectation is consistent with experimental results for $\mathbf{3}^+$.

**Odd-Electron Distribution in $\mathbf{3}^-$.** The odd-electron density on a cobalt $d_{xzi}$ AO, $\rho(\text{Co}, xz)$, of $\mathbf{3}^-$ can be correlated with the cobalt hyperfine splitting tensor with corrections for second-order effects of unquenched orbital angular momenta of electrons:[10]

$$A_{\parallel}(\text{Co}) = a(\text{Co}) + 2(B(\text{Co}, xz))\rho(\text{Co}, xz) + {}^7\!/_6(B(\text{Co}, xz))\Delta g_{\parallel} + {}^1\!/_2(B(\text{Co}, xz))\Delta g_{\perp} \quad (1)$$

$$A_{\perp}(\text{Co}) = a(\text{Co}) - (B(\text{Co}, xz))\rho(\text{Co}, xz) + {}^7\!/_6(B(\text{Co}, xz))\Delta g_{\perp} - {}^1\!/_4(B(\text{Co}, xz))\Delta g_{\perp} \quad (2)$$

where $2B(\text{Co}, xz)$ is the parallel splitting constant of a cobalt nucleus with a unit odd-electron density on its $d_{xzi}$ AO and $a(\text{Co})$ is the Fermi contact term of the cobalt nucleus arising from the spin polarization of the inner-shell s electrons.[11] The valence-orbital parameter of $2B(\text{Co}, xz)$ has been evaluated as $-161 \times 10^{-4}$ cm$^{-1}$.[12,13] By using eq 1 and 2, the Fermi contact term and the odd-electron density on the cobalt d AO are estimated as $a(\text{Co}) = -40 \times 10^{-4}$ cm$^{-1}$ and $\rho(\text{Co}, xz) = 0.18$, respectively. The relatively small odd-electron density on the cobalt d AO implies appreciable mixing of doubly occupied degenerate $\pi$ MO's of the cyclopentadienides and/or cobalt $4p_{xi}$ AO's into the odd-electron orbital.[14]

The odd electron of **1** and $\mathbf{2}^-$ has been shown to be accom-

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(6) Goodman, B. A.; Raynor, J. B. *Adv. Inorg. Chem. Radiochem.* **1970**, *13*, 135.

(7) Doubly occupied $\pi$ MO's of the $C_5H_5^-$ shift $d_{yzi}$, $d_{xzi}$, and $d_{z^2i}$ orbitals upward in the energy diagram.

(8) Walsh, A. D. *Trans. Faraday Soc.* **1949**, *45*, 179.

(9) (a) Kawamura, T.; Enoki, S.; Hayashida, S.; Yonezawa, T. *Bull. Chem. Soc. Jpn.* **1982**, *55*, 3417. (b) Sowa, T.; Kawamura, T.; Shida, T.; Yonezawa, T.; *Inorg. Chem.* **1983**, *22*, 56.

(10) Kawamura, T.; Fukamachi, K.; Sowa, T.; Hayashida, S.; Yonezawa, T. *J. Am. Chem. Soc.* **1981**, *103*, 364.

(11) Note that the odd-electron density on the cobalt 4s AO is zero because of the symmetry of the odd-electron orbital.

(12) Morton, J. R.; Preston, K. F. *J. Magn. Reson.* **1978**, *30*, 577.

(13) (a) Because of Pauling's electroneutrality principle,[13b] we adopt the $2B$ value of Co(0). (b) Pauling, L. *J. Chem. Soc.* **1948**, 1461.

(14) The mixing of Co $4p_{xi}$ AO's into the $a_2'$ odd-electron orbital is expected to be minor, because the odd-electron orbital has antibonding characters with respect to both Co-cyclopentadienide and Co–Co bonds. Since Co 4p AO's are rather diffuse, the mixing of Co 4p AO's into the odd-electron orbital is expected to give little influence on $g$ and hyperfine splitting tensors.

modated in the $a_2$ MO (in the $C_{3v}$ point group),[2,3] which is quasi-equivalent to the odd-electron orbital of **3**$^-$. The odd-electron density on a cobalt d AO can be estimated as 0.24 and 0.25 for **1** and **2**$^-$, respectively, from their spin Hamiltonian tensors[15] listed in Table I. The small cobalt d character of the odd-electron orbital of **3**$^-$ in comparison to those of **1** and **2**$^-$ shows that the cyclopentadienide is a better electron donor than the $(CO)_3$ group.

**g Tensor of 3$^-$ and Orbital Energy Differences.** With use of the theory for the **g** tensor developed by Stone,[16] $g_\parallel$ and $g_\perp$ can be correlated with the electronic structure of **3**$^-$ as in

$$g_\parallel - g_e \simeq 2\zeta|\langle 1a_2'|\sum_i l_{Yi}|1a_1'\rangle|^2/[\epsilon(1a_2') - \epsilon(1a_1')] \quad (3)$$

$$g_\perp - g_e \simeq 2\zeta|\langle 1a_2'|\sum_i l_{Zi}|1e''\rangle|^2/[\epsilon(1a_2') - \epsilon(1e'')] \quad (4)$$

where $\zeta$ is the one-electron $l$–$s$ coupling constant of Co 3d AO's, which is 517 cm$^{-1}$,[17] $l_{Zi}$ is the orbital angular momentum component parallel to the molecular $Z$ axis around the $i$th cobalt nucleus, and $\epsilon$ designates an orbital energy. The MO diagram in Figure 5 together with eq 3 and p 4 redicts $g_\parallel > g_e$ and $g_\perp < g_e$. The prediction is consistent with the experimental results.

If the d characters of 1a′ and 1e″ MO's are assumed to be similar to that of the odd-electron orbital $1a_2'$, the orbital energy differences in the denominators of eq 3 and 4 are estimated as $\epsilon(1a_2') - \epsilon(1a_1') \simeq 1.3$ and $\epsilon(e'') - \epsilon(1a_2') \simeq 1.4$ eV for **3**$^-$.

## Experimental Section

$Co_3(\eta^5\text{-}C_5H_5)_3(\mu_3\text{-}CPh)_2$ was prepared by the method of Fritch and Vollhardt[18] and purified by sublimation (190 °C (0.1 torr)).

**Electrochemistry and $Co_3(\eta^5\text{-}C_5H_5)_3(\mu_3\text{-}CPh)_2^{+\cdot}$ (3$^+$).** Cyclic voltammetry and coulometry experiments were conducted by using a Hokuto-Denko HB-70A function generator and a HB-104A potentiostat/galvanostat and recorded on a Hitachi 057-1001 X-Y recorder.

The cell for electrochemical measurements was composed of two compartments connected by a sintered-glass disk. The main compartment was equipped with a Pt working electrode (area 0.06 cm$^2$) for cyclic voltammetry and an Au working electrode (area 12 cm$^2$) for coulometry. An Au counterelectrode (area 10 cm$^2$) was set in the auxiliary compartment. The reference electrode was SCE, which was isolated from the bulk solution by a sintered-glass disk. To the electrolysis cell was added 10 mL of electrolyte solution, acetonitrile containing 0.1 mol L$^{-1}$ of $n$-$Bu_4NClO_4$, deaerated by Ar bubbling, and then 5.5 mg (1.0 mmol) of **3** was added into the main compartment. The oxidation of **3** was performed at 0.50 V vs. SCE by using the Au working electrode under magnetic stirring until the current reduced below 5% of the initial current. The integration of the current vs. time curve gave 79 C (0.82 × 10$^{-3}$ faraday) as the consumed electricity. Cyclic voltammograms of the oxidized solution were obtained by using the Pt working electrode. When necessary, the oxidized solution was transferred into a quartz sample tube and its ESR spectrum was examined at 77 K. The coulometry for the reduction of the oxidized solution was performed with use of the Au working electrode maintained at the potential of +0.20 V vs. SCE. The amount of electricity consumed for the reduction was 0.72 C (0.75 × 10$^{-3}$ faraday). Examination of the electrolyte solution after the oxidation–reduction cycle by a high-pressure liquid chromatograph equipped with a Cosmosil-5C$_{18}$ column (Nakarai Chemicals Co.) showed that more than 90% of the initial **3** was recovered.

**$Co_3(\eta^5\text{-}C_5H_5)_3(\mu_3\text{-}CPh)_2^{-\cdot}$ (3$^-$).** Standard techniques to prepare ESR samples of anion radicals of aromatic hydrocarbons by alkali-metal reduction[19] was applied to reduce **3** by potassium metal in MTHF. The anion radical was stable in MTHF under vacuum at room temperature and lasted for weeks without detectable changes.

**Measurements and Analysis of ESR Spectra.** X- and K-band ESR spectrometers and monitorings of field sweeps and microwave frequencies have been mentioned earlier.[9a] ESR spectra were simulated with use of a FORTRAN program, SIM(AP),[20] which is based on a second-order perturbational solution of an axially symmetric spin Hamiltonian.[21]

**Acknowledgment.** ESR spectra were simulated on a FACOM M382 computer at the Data Processing Center of Kyoto University.

**Registry No.** **3**, 72271-50-2; **3**$^+$, 87482-30-2; **3**$^-$, 87482-31-3.

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(15) Although the anisotropic spectra of **1** and **2**$^-$ have been tried to fit axially symmetric spin Hamiltonians,[2,3] we expect that the unique axes of the three cobalt splitting tensors for **1** and **2**$^-$ are more or less tilted from the molecular $C_3$ axis. Consequently, the $A_\parallel$(Co) values of **1** and **2**$^-$ in Table I are underestimates, and the odd-electron densities on their Co d AO's in the text should be regarded as estimates of their lower limits.

(16) Stone, A. J. *Proc. R. Soc. London, Ser. A* **1963**, 271.

(17) Dunn, T. M. *Trans. Faraday* Soc. **1961**, *57*, 1440.

(18) Fritch, J. R.; Vollhardt, K. P. C. *Angew, Chem., Int. Ed. Engl.* **1980**, *19*, 559.

(19) Alger, R. S. "Electron Paramagnetic Resonance, Techniques and Applications"; Wiley: New York, 1968; Chapter VI.

(20) Hayashida, S.; Kawamura, T.; Yonezawa, T. *Inorg. Chem.* **1982**, *21*, 2235.

(21) Bleaney, B. *Philos, Mag.* **1951**, *42*, 441.