The parameters of the spin Hamiltonian are obtained experimentally for a paramagnetic square-planar copper(I) complex, and are also derived theoretically using a molecular orbital description of the electronic wave functions. In this way, the electron spin resonance data are used to assess the covalency of the ligand-metal bonds. [The SCI® indicates that this paper has been cited over 335 times since 1961.]

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"I am very pleased that this particular article has been cited sufficiently to qualify it as a Citation Classic. It is based on part of my PhD dissertation research at Berkeley and is the first paper that I ever wrote. I had spent one year as a graduate student from 1952 to 1953 before being inducted into the US Army. On returning to Berkeley from active duty in 1955, I found that a change of direction was called for. This was provided by a new assistant professor who arrived at Berkeley. Bruce McGarvey suggested that I learn about what a new spectroscopic method, electron spin resonance (ESR), could tell us about the nature of bonding in transition metal complexes. McGarvey had recently used ESR measurements on a single crystal of copper(II) bis-acetylacetonate to prove that the unpaired electron is not promoted to a 4p orbital as would be expected on the basis of Pauling's theory of dsp² square bond orbitals. I remember being impressed by McGarvey's explanation that observables such as the metal nuclear hyperfine coupling and the nuclear quadrupole splitting could be related to details of the electronic wavefunction.

"I happily accepted the challenge of finding a quantitative relationship between ESR and chemical bonding in 'covalent' transition metal complexes. I had to find a diamagnetic isomorphous host for the copper(II) bis-acetylacetonate in order to reduce the magnetic interactions and thereby resolve the hyperfine structure. The Pd(II) analog turned out to be an acceptable host. On the theoretical side, I had to learn some quantum mechanics, group theory, and especially molecular orbital theory, which I hoped would be an adequate description for the d-orbital wavefunctions of covalent complexes. Among the most difficult but also most enjoyable parts of the problem was learning the classic theoretical papers of Pryce and Abragam. My contribution was to extend this theory—originally written for ionic complexes—to covalent complexes with extended molecular orbital wavefunctions. McGarvey helped out at critical times when I was bogged down but by and large he left me alone while working on his own theoretical projects in paramagnetic relaxation.

"What factors do I think contributed to the relatively large number of citations? In 1958, when our paper was published, ESR was in the process of passing from the hands of physicists, who invented it, to chemists. Simultaneously, the first commercial ESR spectrometers were becoming available. The next 15 to 20 years produced a tremendous number of papers dealing with ESR spectroscopy, particularly from chemists. (For more recent developments see, for instance, Electron Spin Resonance and references contained therein.) Our paper presented a timely formulation of ESR experimental parameters of a transition metal complex in the context of molecular orbital theory and bond covalency, which are familiar concepts to most chemists. Thus, for many chemists, it was of use in discussing their experimental data."