The Kubo linear-response formalism is applied to the study of electron spin resonance linewidths of free radicals in liquid solutions. Simple formulas describing these linewidths in terms of magnetic parameters and the rotational correlation time are given. [The *SCI®* indicates that this paper has been cited in over 590 publications since 1960.]

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April 19, 1985

Electron spin resonance (ESR) studies of free radicals in liquid solutions date from the early 1950s. At first, interest focused on determining hyperfine splittings and g-factors and using these magnetic parameters in probing spin density distributions in free radicals. Soon thereafter attention was given to the spectral linewidths, first because they had to be understood to interpret the spectra, and later because they could be used to study molecular relaxation in liquids.

In 1956, while I was spending the summer at Columbia University, George Fraenkel suggested that I read the article by Kubo and Tomita[1] in which a powerful formalism was presented for handling spin relaxation. I was a slow learner, and it took me much time, but I recognized that with this formalism the problem of ESR linewidths could be described in a very straightforward manner. And this is what I attempted to do in the article cited.

The article describes many sources of linebroadening but focuses on exchange and anisotropic magnetic properties—hyperfine interactions and g-tensors—and the asymmetry of the spectrum. It was not until later that Peter Atkins and I became aware that spin-rotational effects could also contribute significantly to ESR linewidth.[2]

Earlier work on linewidths had already underscored the relevant physics, in particular, articles on asymmetric spectra by McGarvey[3] and by McConnell[4] and on spin exchange by Tuttle and Pake.[5] These articles told me how to begin and what to expect. But with the Kubo formalism, one could study linewidths without the use of ingenuity—all one needed was a reasonable spin Hamiltonian. The success of the article, I believe, lies in just this fact—that workers in the field can readily understand the discussion of linewidths and can, without difficulty, make use of the results in analyzing their ESR spectra. Perhaps this is why researchers kindly still refer to the article even though more powerful and complete studies have since been presented.[6]

1. **Kubo R & Tomita K.** A general theory of magnetic resonance absorption. *J. Phys. Soc. Jpn.* 9:888-919. 1954. (Cited 1,160 times since 1955.)
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3. **McGarvey B R.** Paramagnetic resonance in copper chelates. *J. Phys. Chem.* 60:71-6. 1956. (Cited 110 times.)
4. **McConnell H M.** Effect of anisotropic hyperfine interactions on paramagnetic relaxation in liquids. *J. Chem. Phys.* 25:709-11. 1956. (Cited 350 times.)
5. **Pake G E & Tuttle T R. Jr.** Anomalous loss of resolution of paramagnetic resonance hyperfine structure in liquids. *Phys. Rev. Lett.* 3:423-5. 1959. (Cited 105 times.)
6. **Freed J H & Fraenkel G K.** Theory of linewidths in electron spin resonance spectra. *J. Chem. Phys.* 39:326-48. 1963. (Cited 435 times.)