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NOTES

Comment on "2D NMR Nutation Spectroscopy in Solids" by A. Samoson and E. Lippmaa

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In a recent paper (*1*), Samoson and Lippmaa present theoretical and numerical calculations to aid in the interpretation of experimental nutation spectra. Their paper presents a number of cases of 1/2-integral spin results based upon the on-resonance response of the spins to the Hamiltonian in the rotating frame,

$$\mathscr{H} = -\omega_{\mathrm{RF}} I_X + \omega_{\mathrm{Q}}(3I_Z^2 - I^2) \qquad [1]$$

The purpose of this Note is to point out a misleading and incorrect statement found at the top of page 257. The authors use a density matrix approach based upon a representation using the eigenfunctions $|m\rangle$ of I_Z,

$$|\pm m\rangle = (|m\rangle \pm |-m\rangle)/\sqrt{2}. \qquad [2]$$

They refer to the spherical tensor approach of mine (*2, 3*) but state, however, "The new formalisms (spherical tensors) have been designed to evaluate the total single-quantum dipolar coherence as an observable. This is not suitable for nutation spectroscopy, where the central transition coherence is observed as a sum of contributions from all possible multiple coherences." These statements are incorrect.

First, the spherical tensor approach, called multipole NMR, was developed in order to specifically treat NMR multiquantum coherences. It does this naturally since the spherical tensor component, q, is equal to $\Delta m = m' - m$. As such it describes the multiquantum coherence. The papers referenced by Samoson and Lippmaa (*2, 3*) as well as others (*4*) make this point.

Second the NMR observable is the magnetization, proportional to the total single-quantum dipolar coherence. This, however, is not the only observable of interest since many multiquantum coherences can be indirectly observed. These have been calculated, for example, (*5, 6*), and their existence confirmed experimentally (*5*). The multipole formulation was developed in order to describe polarizations such as these having both multipole character and multiquantum coherence.

Nutation spectroscopy can also be treated within the multipole formalism, as can any spin observable. Let $\hat{I}_{ij}$ correspond to a fictitious spin-1/2 operator. This may well describe the central line, or any other specific multiquantum frequency. Since the

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multipole approach evaluates the spin density operator $(\rho(t))$ the expectation value solves the problem,

$$\langle \hat{I}_{ij} \rangle = \mathrm{Tr}\{\hat{I}_{ij}\rho(t)\}. \qquad [3]$$

Evaluation of the trace is no more difficult, once the density matrix is found, when the density matrix is represented in a spherical tensor basis or in the basis used in Ref. (*1*), [2].

Finally, any convenient basis can be used to evaluate the spin density matrix. Some lend themselves more to approximations than others but for numerical calculations there is little advantage of one basis over another.

The spherical tensor approach is not new and has wide applications in areas such as nuclear physics, spectroscopy, transport phenomena, and scattering theory. Phenomena which display rotational symmetry usually benefit from a spherical tensor formalism (*7*).

REFERENCES

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7. See, e.g., E. P. Wigner, "Group Theory," Academic Press, New York, 1959, or L. C. Biedenharn and J. D. Louck, "Angular Momentum in Quantum Physics," Addison–Wesley, Reading, Massachusetts, 1981.