Comments on Effects of Electron-Electron Interactions on Nuclear Spin-Lattice Relaxation Times in Aluminum - A Reply

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The comments by Fradin and Rowland[1] (referred to as FR) on our paper[2] on the effects of electron-electron interactions in aluminium on the nuclear relaxation times therein are concerned primarily with our estimate of the size of the quadrupole baths in high-purity aluminium. As mentioned by FR, many observers have noted the anomalous width of the aluminium nuclear-resonance line and several have suggested quadrupole effects as the cause. The observation[3] of strong spin absorption in zero field at frequencies as high as 80 kHz in 99.9999% pure aluminium is the latest indicator of strong quadrupole effects in the ostensibly pure metal. We have simulated this quasicontinuous distribution of quadrupolar splittings by two baths, with strong internal coupling and weak interactions (a) with each other, and (b) with the dipolar bath, a logical extension of models widely utilized by other workers.[4]

We arrived at our estimates of the heat capacities of these baths in our sample by fitting the experimental results on the measurement of apparent dipolar relaxation times to the predictions of a three-bath model with a dipolar, plus two quadrupolar baths. The estimate of the smaller quadrupolar bath was supported by the observation of a nonexponential decay at helium temperatures, with the more rapid component of the decay having a time constant ~ 20 msec and an amplitude consistent with the presence of a quadrupolar bath of heat capacity one-half of that of the dipolar bath. The theoretical model is rudimentary, being the simplest which would approach an explanation of our measurements. Each bath is assumed to be characterized by internal equilibrium at the start of our measurements and we postulate that our experimental technique, although measuring the temperature of the dipolar bath as a function of time, will lead to the observation of a complex nonexponential decay reflecting the weak thermal coupling of the dipolar bath to the lattice and to the first quadrupolar bath, as well as the coupling between the two quadrupolar baths.

This approach is fundamentally different to that adopted by FR where the basic assumption is, to quote, "that cross relaxation is sufficiently fast to establish a common spin temperature for the Zeeman, dipolar, and quadrupolar systems." In particular the FR estimate of the size of the quadrupolar bath is based on the magnetization amplitude immediately after the spin-locking pulse sequence, i.e., a few tens of microseconds after the pulses establishing the nonequilibrium magnetization in the rotating frame. The comparison between the two sets of experiments is further complicated by the presence of a rotating field during the FR experiments. It is most unlikely that the widely different techniques would lead to comparable estimates of the size of any quadrupolar effects present in high-purity annealed metal. We reiterate here the purity specification of our sample; a resistance ratio of 3650 and a measured second moment of 12.3 kHz,[2] fully consistent with the measurements on 99.9999% purity Al quoted by Fernelius.[4]

The value of δ that FR quote as measured by their technique, 2.65, as compared to our value of 2.15, is more worrying. It seems to us, from our arguments in the preceding paragraph, that the size of the quadrupole bath in thermal contact with the dipolar bath in the FR measurements will vary (a) during a single measurement, at fixed H_1,

Note added in proof. The absence of an error estimate on the FR value of $\delta = 2.65$ is disturbing. The data, as presented in Fig. 1 of Ref. 1, do indicate that a substantial error bar is needed.

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[1]F. Y. Fradin and T. J. Rowland, preceding paper, Phys. Rev. B 3, 1781 (1971).

[2]D. P. Tunstall and D. Brown, Phys. Rev. B 1, 2881 (1970).

[3]M. Minier, Phys. Rev. 182, 437 (1969).
[4]N. Fernelius, in *Proceedings of the International Conference on Magnetic Resonance and Relaxation, Fourteenth Colloque Ampère, Ljubljana, Yugoslavia, September*, 1966, edited by R. Blinc (North-Holland, Amsterdam, 1968), p. 497.
[5]J. H. Pifer, Phys. Rev. 166, 540 (1968).