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The paper on "Solid-State 1H Spin Polarimetry by 13CH3 Nuclear Magnetic Resonance" by Stuart J. Elliott, Quentin Stern (or should it be Quentin Chappuis, as in the Supplement?) and Sami Jannin raises all sorts of interesting questions.

When the authors write “The frequency of the dashed line corresponds to the minimum between the two peaks at high levels of 1H polarization,” they discuss the two peaks phenomenologically as if they constituted a doublet. Are they implicitly referring to a doublet due to the heteronuclear dipole-dipole coupling in the Hamiltonian?

Is this splitting only seen in methyl groups? Do these methyl groups have to rotate freely all the way down to sample temperatures near 1.2 K? Is that the case for methyl groups in acetate? Do you see similar effects in molecules such as gamma picoline that are known to have very low rotation barriers? If the methyl rotation in acetate is frozen, why are the methyl signals in acetate any different from those of other rigid chemical moieties like CH2 groups? You suggest that similar effects can be observed in molecules “such as [1-13C]sodium formate”, although there are no methyl groups in formate.

Can you explain your observation “where the [proton] polarization is higher, the peak intensities become more equal”? Can you explain why "[proton] polarizations (…) were observed to *decrease* linearly with *increasing* 13C peak asymmetries”? As you wrote, this behaviour is opposite to what was observed by Mammoli et al. and by Aghelnejad et al. It seems that polarisation and asymmetry should increase together.

What do you expect to see in the presence of “solid-state methyl group AE population imbalances at low temperatures”?

For practitioners of DNP, it would be nice to know more details about how you determined the "power of ca. 80 **P**μw = 125 mW at the output of the microwave source and ca. **P**μw = 30 mW reaching the DNP cavity (evaluated by monitoring the helium bath pressure.)” Is the first number given by the manufacturer of the microwave source? How did you convert pressure changes into mW? How do you calibrate this empirical relationship?

The statement “the 1H and 13C [transverse] relaxation time constants in the presence of an rf-field are extended by orders of magnitude” seems a bit exaggerated. Bornet et al. have shown experimentally that T_1rho can be extended by a factor 5 or so when the microwaves are switched on.

I hope that these naive comments will contribute to make the “discussions” of “Magnetic Resonance” more lively, and may help to reduce inhibitions and “open the gates” to further comments from interested readers.