Comment on "^{13}C NMR Study of Layered Organic Superconductors Based on BEDT-TTF Molecules"

In the recent Letter [1], Kawamoto *et al.* reported the study of ^{13}C NMR line shape and relaxation versus temperature, performed on powdered samples of several BEDT salts. The authors could have attempted to perform a comparison of their results with a study carried previously on a single crystal of κ-(ET)$_2$Cu[N(CN)$_2$]Br [2]. The purpose of this Comment is to clarify some ambiguities in the interpretation of their data and to show that some of their conclusions are incomplete or misleading.

The first comment addresses the determination of the Knight shift tensor presented by the authors. The theoretical calculation gives the hyperfine field at the central carbon site as a sum of dipolar on-site and off-site contributions. This calculation gives qualitatively correct results such as a nearly uniaxial symmetry of the tensor and the correct order of magnitude of the hyperfine coupling, however, the experimental data do not permit a fine adjustment of theoretical parameters, since it is merely based on the powder line shape analysis. On the other hand, the data published in [2] were obtained from a *direct* measurement of the Knight shift tensor by fitting the single crystal rotation patterns completed by other NMR experiments used for the line assignment. We remind one here of the following conclusions of this study: (1) The two central carbon sites of ET in κ-(ET)$_2$Cu[N(CN)$_2$]Br are not equivalent due to the dimerization of the BEDT stack. The main tensor elements differ by 150 ppm or so, which cannot be neglected compared to the shift anisotropy ($\approx$300 ppm) and gives an important contribution to the powder-averaged linewidth. The model in the Ref. [1] could only work with different spin densities for each C and S site. The same arguments hold for the calculation of the "structure factor" of the relaxation rate. (2) The asymmetry between the shifts of the two carbons of the C$=$C bond proves that the influence of adjacent BEDT molecules on the hyperfine field is important. Therefore the hypothesis made by the authors about the principal axes of the K tensor is not obvious.

Furthermore, the discussion of the temperature dependence of the susceptibility presented by the authors has to be revised. As we showed in [2], the Knight shift shows a substantial decrease below 50 K, also followed by the ESR susceptibility [3]. This effect is absent in [1] because of an additional source of broadening due to a lattice distortion [2,4] (see Fig. 1). Therefore, studying the powder linewidth, the authors missed the important result which is the possible existence of a pseudogap below the maximum of $1/T_1T$.

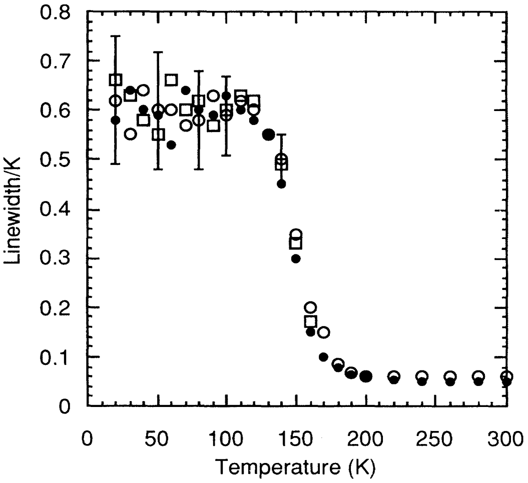

FIG. 1. Linewidth normalized to the Knight shift for several lines in the spectrum.

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