Comment on “Low Temperature Magnetic Instabilities in Triply Charged Fulleride Polymers”

Recently, Arçon et al. \cite{1} reported ESR studies of the polymer phase (PP) of Na$_2$Rb$_{0.3}$Cs$_{0.7}$C$_{60}$ fullerite. It was claimed that this phase is a quasi-one-dimensional metal above 45 K with a spin-gap below this temperature and has antiferromagnetic(AF) order below 15 K, that is evidenced by antiferromagnetic resonance(AFMR).

For the understanding of the rich physics of fullerides it is important to identify the different ground states. ESR has proven to be a useful technique for this purpose. However, since it is a very sensitive probe, it can detect a multitude of spin species and it is not straightforward to identify their origin, especially in a system like Na$_2$Rb$_2$Cs$_{1-x}$C$_{60}$ with three dopants, when one part of the sample polymerizes but the majority does not.

The observation of a low dimensional instability in the single bonded PP would be a novel and important result. Nevertheless, in this Comment we argue that Na$_2$Rb$_{0.3}$Cs$_{0.7}$C$_{60}$ is not a good choice for this purpose since, as we show, the samples used in Ref. \cite{1} are inhomogeneous. We point out that recent results on the PP of Na$_2$CsC$_{60}$ contradicts the observation of low dimensional instabilities in Na$_2$Rb$_{0.3}$Cs$_{0.7}$C$_{60}$.

The ESR signal of Ref. \cite{1} at 285 K consists of two components as read from Fig. 1 of Ref. \cite{1}. For the narrower component the peak-to-peak linewidth $\Delta H_{pp}$ $\approx$ 60 G. This is not the signal of the simple cubic(sc) phase of Na$_2$Rb$_{0.3}$Cs$_{0.7}$C$_{60}$ as $\Delta H_{pp}$ $\approx$ 250 G is expected for this phase from measurements on the sc phases of Na$_2$CsC$_{60}$ ($\Delta H_{pp}$ $\approx$ 370 G \cite{2}) and Na$_2$RbC$_{60}$ ($\Delta H_{pp}$ $\approx$ 40 G \cite{3} \cite{4}) at 285 K. A broader component in the ESR signal in Fig. 1 of Ref. \cite{1} is indeed visible, as a tail around 0.3 T and is better seen in the integrated spectrum at 220 K with $w$ $\approx$ 200 G ($\Delta H_{pp}$ $\approx$ 230 G). This multi-component nature of the ESR signal suggests a phase separation rather than local disorder as the latter would be averaged by conduction electrons. The narrow(broad) ESR component may come from Rb(Cs) rich and Cs(Rb) poor parts of the sample. The non-stoichiometry of the compound may be the reason for this phase separation that affects the PP, as well. It is not documented in Ref. \cite{1} whether contrast between Rb and Cs in the X-ray experiment allows the exclusion of the above suggested phase separation.

In addition to the apparent phase separation of the sample used in the experiment, we could not reproduce the temperature dependence of the ESR intensity in Fig. 3a and c from the raw ESR spectra of Fig. 2 of Ref. \cite{1} and from the $w_T$ values of Fig. 3b and d. It appears as if the broadening of the ESR line below 45 K (that reduces the amplitude of the ESR signal of the powder distribution) was not taken into account of the calculation of the ESR intensity. This leads us to question the observation of a spin-gap below 45 K.

Below 15 K, Arçon et al. \cite{1} attributes the AFMR to an emerging ESR signal in high frequency (HF)-ESR which is absent in X-band. The HF-ESR linewidth and field shift from the ESR signal of the PP allowed the calculation of a reasonable value of spin-flop(SF) field. This is insufficient for the unambiguous identification of an AFMR, that requires ESR measurements at least at two high frequencies above the SF field \cite{5} and the observation of a decreasing linewidth and field shift with increasing ESR frequency. It would be important to see the result of these experiments that could be performed at NHMFL \cite{6}.

In our opinion, it is very intriguing that the presence of superconducting phase in the sample is neglected in the discussion of Ref. \cite{1}. 81 % of Na$_2$Rb$_{0.3}$Cs$_{0.7}$C$_{60}$ is in the sc phase and is a superconductor with $T_c$ $\approx$ 10 K \cite{6}. The so called vortex noise due to this significant amount of superconducting phase probably prevents reliable conclusions from X-band data (not shown in Ref. \cite{1}) below $T_c$. Thus it can not be decided whether the signal that is observed in the HF experiment is present or not in X-band. Moreover, it can not be excluded that the signal observed below 15 K in the HF experiment and attributed to the AFM phase is the ESR signal of the residual $sc$ phase. We estimate that $w$ $\approx$ 7 mT for the $sc$ phase at 15 K from results on Na$_2$CsC$_{60}$ \cite{2} and Na$_2$RbC$_{60}$ \cite{1}. This ESR line is expected to narrow below $T_c$ similarly to the situation encountered in K$_3$C$_{60}$ \cite{8}, which may lead to a linewidth of the $sc$ phase similar to that assigned to the AFMR signal.

In summary, we have shown that the analysis and discussion of the experimental data in Ref. \cite{1} is ambiguous and full reconsideration of the results is necessary. The main reason of the authors of Ref. \cite{1} to expect electronic instabilities in the PP of Na$_2$Rb$_{0.3}$Cs$_{0.7}$C$_{60}$ is the expanded interchain distance in comparison with e.g. the PP of Na$_2$RbC$_{60}$, the latter being a metal till 4 K. It has recently been shown that the even further expanded lattice size Na$_2$CsC$_{60}$ polymer \cite{9} \cite{10} is a metal until low temperatures \cite{2}. Thus, the ground state of the single bonded fulleride polymers is more likely a metal than a spin density wave insulator.

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