Comment on "Low Temperature Magnetic Instabilities in Triply Charged Fulleride Polymers"

Recently, Arčon *et al.* [1] reported ESR studies of the polymer phase (PP) of $Na_2Rb_{0.3}Cs_{0.7}C_{60}$ fulleride. 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 (AFM) 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 such as $Na_2Rb_xCs_{1-x}C_{60}$ 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 $Na_2Rb_{0.3}Cs_{0.7}C_{60}$ is not a good choice for this purpose since, as we show, the samples used in Ref. [1] are inhomogeneous. We point out that recent results on the PP of Na_2CsC_{60} contradicts the observation of low-dimensional instabilities in $Na_2Rb_{0.3}Cs_{0.7}C_{60}$.

The ESR signal of Ref. [1] at 285 K consists of two components as seen in Fig. 1 of Ref. [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 $Na_2Rb_{0.3}Cs_{0.7}C_{60}$ as $\Delta H_{pp} \approx 250$ G is expected for this phase from measurements on the sc phases of Na_2CsC_{60} ($\Delta H_{pp} \approx 370$ G [2]) and Na_2RbC_{60} ($\Delta H_{pp} \approx 40$ G [3]) at 285 K. A broader component in the ESR signal in Fig. 1 of Ref. [1] is indeed visible, as a tail around 0.3 T and is better seen in the integrated spectrum at 200 K with $w \approx 200$ G ($\Delta H_{pp} \approx 230$ G). This multicomponent 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 nonstoichiometry of the compound may be the reason for this phase separation that affects the PP, as well. It is not documented in Ref. [1] whether contrast between Rb and Cs in the x-ray experiment allows the exclusion of the above suggested phase separation.

In addition, we could not reproduce the T dependence of the ESR intensity in Figs. 3a and 3c from the raw ESR spectra of Fig. 2 of Ref. [1] and from the w_T values of Figs. 3b and 3d. It appears as if the broadening of the ESR line below 45 K was not taken into account in the calculation of the ESR intensity. This leads us to question the observation of a spin gap below 45 K.

Below 15 K, Arčon *et al.* [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, which requires ESR measurements at least at two high frequencies above the SF field [4] and the observation of a decreasing linewidth and field shift with increasing ESR frequency.

It is very intriguing that the presence of superconducting (S) phase in the sample is neglected in the discussion of Ref. [1]. 81% of $Na_2Rb_{0.3}Cs_{0.7}C_{60}$ is in the sc phase and is a superconductor with $T_c \approx 10$ K [5]. The so-called vortex noise due to this significant amount of S phase probably prevents reliable conclusions from X-band data (not shown in Ref. [1]) below T_c. Thus it cannot be decided whether the signal that is observed in the HF experiment is present or not in X-band. Moreover, it cannot 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_2CsC_{60} [2] and Na_2RbC_{60} [3]. This ESR line is expected to narrow below T_c similarly to the situation encountered in K_3C_{60} [6], 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. [1] is ambiguous and full reconsideration of the results is necessary. The main reason for the authors of Ref. [1] to expect electronic instabilities in the PP of $Na_2Rb_{0.3}Cs_{0.7}C_{60}$ is the expanded interchain distance in comparison with, e.g., the PP of Na_2RbC_{60}, the latter being a metal till 4 K. It has recently been shown that the even further expanded lattice size Na_2CsC_{60} polymer [7,8] is a metal until low T's [2]. Thus, the ground state of the single bonded fulleride polymers is more likely a metal than a spin density wave insulator.

Ferenc Simon, Slaven Garaj, and László Forró
Département de Physique
École Polytechnique Fédérale de Lausanne
CH-1015, Lausanne, Switzerland

Received 9 August 2000; published 29 August 2001
DOI: 10.1103/PhysRevLett.87.129703
PACS numbers: 75.30.Fv, 61.48.+c, 76.30.−v

[1] Denis Arčon *et al.,* Phys. Rev. Lett. **84**, 562 (2000).
[2] N. Čegar *et al.*, cond-mat/0002447.
[3] Denis Arčon *et al.,* Phys. Rev. B **60**, 3856 (1999).
[4] A. Jánossy *et al.,* Phys. Rev. Lett. **79**, 2718 (1997).
[5] T. Yildirim *et al.,* Solid State Commun. **93**, 269 (1995).
[6] N. M. Nemes *et al.,* Phys. Rev. B **61**, 7118 (2000).
[7] The lattice constants of $Na_2Rb_{0.3}Cs_{0.7}C_{60}$ were not presented in Ref. [1].
[8] S. Margadonna *et al.,* J. Solid State Chem. **145**, 471 (1999).

