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Magnetism of α- and β-TDAE-C_{60}

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Abstract

α-TDAE-C_{60} is a purely organic soft ferromagnet with $T_C \approx 16$ K. The mechanism of the ferromagnetism was recently proposed to be antiferro-orbital-ordering of Jahn–Teller distorted C_{60}^- (Mizoguchi et al., Phys. Rev. B 63 (2001) 040417(R)), which reproduces the pressure dependence of T_C. Further, [2 + 2]-polymerized β-TDAE-C_{60} was found under pressure more than 9 kbar, where the spin on TDAE missing in α-phase revives. This paper reports on the effect of uniaxial strain to T_C, an increase up to 17.5 K, and on the nature of the polymeric β-TDAE-C_{60}.

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Keywords: TDAE-C60; ESR; Organic ferromagnet; Fulleride; Pressure

Organic ferromagnet with a high T_C of 16 K found in α-TDAE-C_{60}, where TDAE is tetrakis-dimethylamino-ethylene, has attracted a lot of interest in its mechanism [1]. Although a variety of mechanisms for the ferro-magnetism have been proposed, the most probable one might relate to the antiferro-orbital-ordering of Jahn–Teller distorted C_{60} balls at the moment [1–3]. The origin of the observed ESR signal in α-TDAE-C_{60} has been assigned to the spin on C_{60}^- as confirmed by the *g*-factor close to that of C_{60}^- radical, as presented in Fig. 1. Remarkably, the *g*-factor in polymeric β-TDAE-C_{60} is in between those for the same [2 + 2] polymer of RbC_{60} and $TDAE^+$ radical, which strongly suggests a revival of the spin in $TDAE^+$, well consistent with the number of spins determined by SQUID susceptometer at ambient pressure, shown in Fig. 2. Since the β-phase is a quasi-stable state at ambient pressure up to 430 K [2], a long-term decreasing drift of the *g*-factor was observed by −0.0005 for 3 months [5]. A possible contribution of the spin in $TDAE^+$ to the ferromagnetism has been a key point in its mechanism. However, a role of the spin in $TDAE^+$ is not clear at this point, since the β-phase is paramagnetic substance.

The first preliminary data on the effect caused by the uniaxial strain along *b*-axis, the direction reducing the distance between C_{60} and TDAE, was demonstrated in Fig. 3. A definite enhancement of T_C was observed up to 17.5 K, the highest T_C, in our knowledge, observed in the pure organic ferromagnets, so far. The strain of *b*-axis does not change the nearest neighbor distance (*c*-axis) or does only a little the 2nd nearest one of C_{60}, but might strongly enhance the anion–cation interaction. Following the antiferro-orbital-ordering model [1–3], this strain would produce only a weak but similar effect to that of hydrostatic pressure [1] which reduces the cancellation of the transfer integral between the ground states, giving rise to a weak enhancement of the negative exchange coupling between the neighboring columns. Then, a possible contribution of $TDAE^+$ to the ferromagnetism to account for the observed increase of

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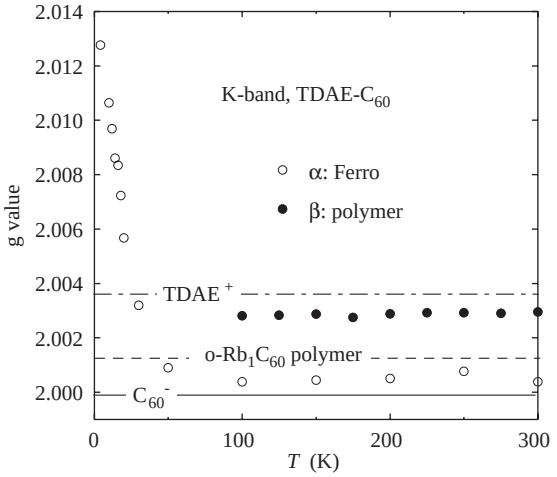

Fig. 1. *g*-factors both for the ferromagnetic α- and the polymeric β-TDAE-C_{60}. Note that the former locates vicinity of 1.9998 for C_{60}^- radical and the latter does in between 2.0036 for $TDAE^+$ radical and 2.0012 for RbC_{60} [2 + 2] polymer [1,4].

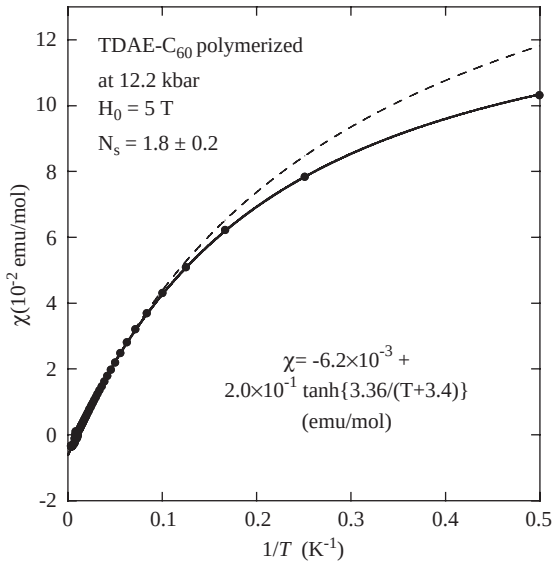

Fig. 2. Reproduction of the magnetic susceptibility of β-TDAE-C_{60} in terms of Brillouin function with a small Weiss temperature (solid line). Almost two spins per each pair of $TDAE^+$ and C_{60}^- were observed, suggesting a revival of the spin in $TDAE^+$. Broken line represents Curie–Weiss law fitted to the high temperature data above 10 K.

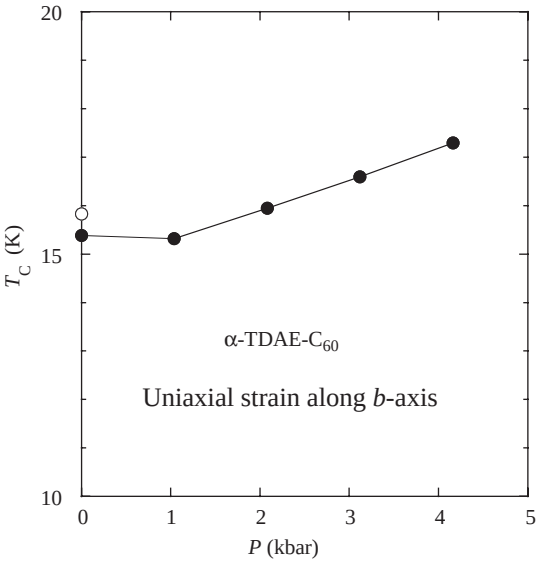

Fig. 3. The effect of uniaxial strain along *b*-axis. Note the increase of T_C up to 17.5 K at 4 kbar. The open circle was taken at ambient pressure, while the closed circles were in the stycast 2850FT which produces additional isotropic pressure to the sample in some extent. Above 3 kbar, two phases appeared with $T_C \approx 10$ K and that shown in this figure, which suggests a delicate balance of interactions as the mechanism of the ferromagnetism.

T_C would be expected. The uniaxial strain dependence of the *g*-factor and T_C along the three directions are crucial to confirm this point and to understand this system further.

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