# Antiferromagnetic ordering in organic $\pi - d$ hybrids [Pd(tmdt)$_2$]

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Emergent phenomena in the systems of interacting electrons are expected to be more diverse when electrons are accommodated by multi-orbitals with distinctive characters. [$M$(tmdt)$_2$], a family of organic $\pi-d$ hybrids, are multi-orbital correlated electron systems, where a transition metal ion, $M$, is coordinated with organic ligands, tmdt, from both sides.[1–3] The orbitals near the Fermi level are a $d$-orbital in $M$ and $\pi$-orbitals in tmdt.[4,5] The energy-level difference between the $d$ and $\pi$ orbitals depends on $M$, and leads to the diverse ground states; paramagnetic metals with appreciable electron correlations for $M$ = Ni and Pt, an antiferromagnetic (AF) metal ($T_N$ = 110 K) for $M$ = Au, and a one-dimensional AF Mott insulator ($T_N$ = 13 K) for $M$ = Cu.[6–11]

For the $M$ = Ni, Pt, and Pd compounds, the $\pi$ and $d$ orbitals are well separated in terms of energy; their conduction bands are composed of only $\pi$ orbitals. Although the Ni and Pt systems are paramagnetic metals as expected, we recently found that the Pd system was exceptional. It showed a decrease in ESR signal intensity and broadening of the NMR spectra below 100 K.[12,13] A broad peak appeared around 50 K in the temperature dependence of the NMR relaxation rate.[13] These results suggest that an AF ordering inhomogeneously appears in the sample. The inhomogeneously-ordered state might be a long-range-ordered state, which cannot be explained by the band calculation and implies the importance of the electron correlation of the $\pi$-orbitals and/or spin-orbit coupling in this system.

In order to achieve full confirmation of the appearance of magnetic ordering and to investigate the magnetic ground state, we carried out zero-field (ZF) $\mu$SR measurements on [Pd(tmdt)$_2$] polycrystalline sample in the DOLLY area at Paul Scherrer Institut. Figure 1(a) shows the ZF-$\mu$SR time spectra of [Pd(tmdt)$_2$] at various temperatures. It can clearly be seen that the relaxation becomes faster at lower temperatures and that the shape of the relaxation function changes with temperature. At 50 K and 10 K, muon-spin precession signals were observed. This provides clear evidence for the appearance of a long-range-ordered magnetic state in [Pd(tmdt)$_2$] at least below 50 K. The slow relaxation of the non-oscillating signal can originate from tempo-

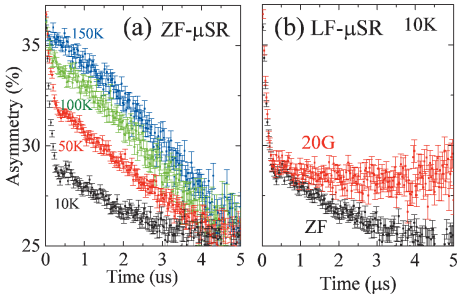


Fig. 1. (a)ZF-$\mu$SR time spectra of [Pd(tmdt)$_2$] observed at 10, 50, 100, and 150 K. (b) LF-$\mu$SR time spectra observed at 10 K.

rally fluctuating fields and/or small static local fields. To determine whether the spin state in the ground state is static or not and to find the residual spin dynamics, we performed $\mu$SR measurements under a longitudinal field (LF). As shown in Fig. 1(b), the slowly relaxing signal was suppressed and became nearly flat on applying a field of 20 G. This indicates the existence of a small static field, thus ruling out the temporally fluctuating fields that are slower than the $\mu$SR time scale ($10^{-6}$–$10^{-11}$ sec). The small static field might originate from the nuclear dipolar fields in a possible paramagnetic volume within the sample. This suggests that the magnetically-ordered state of [Pd(tmdt)$_2$] is a inhomogeneously-ordered state, which is consistent with the preceding NMR and ESR results.

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