Measurement of interlayer spin diffusion in the organic conductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl

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Electron hopping between neighboring layers in quasi-two-dimensional conductors is blocked if incoherent momentum scattering within the layers is strong [1]. The interlayer tunneling hopping rate is $\gamma_{\alpha\beta} = 2t_\perp^2\tau/\hbar^2$ where $t_\perp$ is the overlap integral between layers and τ the in-plane momentum life time. In layered organic conductors $t_\perp$ is small, and at high temperatures, τ is short and a very effective blocking of transport perpendicular to the layers is expected. Although estimates of the anisotropy between in- and out-of-plane conductivities for the κ-(BEDT-TTF)$_2$-X family based on the blocking mechanism exceed 10^5 at ambient temperatures, anisotropies measured by dc methods are orders of magnitude smaller.

Here we measure the interlayer hopping rate quantitatively with a novel method [2]. We find that the perpendicular conductivity derived from the transverse hopping rate is extremely low and is compatible with the expected very large conductivity anisotropy. The method is based on the measurement of interlayer spin diffusion. We observe the motional narrowing of the conduction electron spin resonance (CESR) lines of adjacent layers with slightly different g-factor anisotropies. Using ESR spectroscopy at multiple frequencies in the mm-wave range, we measure the interlayer hopping rate in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl at 250 K as a function of pressure and as a function of temperature at a few selected pressures up to 10 kbar. We find that at ambient pressures and 250 K electrons are confined to a single molecular layer for 3×10^{-9} s. Electrons diffuse within an approximately 1 micrometer diameter circle without hopping to an adjacent layer. The hopping rate increases rapidly under pressure at 250 K. At ambient pressure interlayer hopping is slow at all temperatures in the metallic phase. At higher pressures $\gamma_{\alpha\beta}$ increases with decreasing temperatures. A comparison to in-plane resistivity measured under conditions similar to the ESR study shows that deconfinement of electrons is to a large extent due to an increase of the in-plane momentum life time as predicted by Kumar and Jayannavar [1].

*This work was supported by the Hungarian National Research Fund NK60984, PF63954, K68807.
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