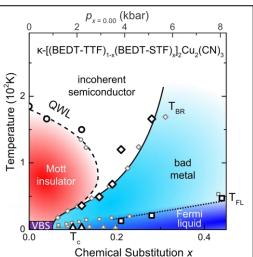
κ-(BEDT-TTF)₂Cu₂(CN)₃: From Spin Liquid to Fermi Liquid

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κ-(BEDT-TTF)₂Cu₂(CN)₃ is among the best studied correlated electron systems and a model compound in many regards [1]. The triangular arrangement of dimers causes strong frustration with no magnetic order despite the sizeable interactions. Our broadband ultra-low-temperature ESR spectroscopy enabled us to unambiguously reveal a rapid drop of the spin susceptibility at the enigmatic 6 K anomaly. This opening of a spin gap, accompanied by structural modifications, evidences a valence-bond-solid ground state. We identify an impurity contribution to the ESR response that becomes dominant when the intrinsic spins form singlets [2].

When the electronic bandwidth is tuned either by external pressure or by chemical substitution, κ -(BEDT-TTF)₂Cu₂(CN)₃ crosses the first-order insulator-metal transition below $T_{\rm crit} \approx 20$ K, where metallic and insulating regions coexist spatially, causing a divergency in the dielectric permittivity with $\epsilon_1 \approx 10^5$ and more [3].



Chemical Substitution xPhase diagram of κ -(BEDT-TTF)₂Cu₂(CN)₃. For strong correlations, the highly frustrated Mott insulator enters a valence bond state below 6 K. On the metallic side – above the Fermi liquid state – a bad-metallic regime occurs where a displace Drude peak is observed in the optical spectra (after [1,4]).

On the metallic side, the low-energy properties can be understood in terms of long-lived Landau quasiparticles with all complex interactions included in Fermi-liquid parameters, such as the effective mass m^* and scattering rate $1/\tau$. Here we investigate the spectral and temperature-dependent complex electrodynamic response $\sigma_1(\omega,T) + i\sigma_2(\omega,T)$ as the effective correlations U/W are varied by chemical tuning. We reveal a persistent Fermi-liquid behavior with T^2 and ω^2 dependences of the optical scattering rate $1/\tau$. The strong increase of the resistivity beyond the Ioffe-Regel-Mott limit $\rho \gg \rho_{\rm IRM}$ is accompanied by a displaced Drude peak in $\sigma_1(\omega)$. Our results, supported by a theoretical model for the optical response, demonstrate the emergence of a bad metal from resilient quasiparticles that are subject to dynamical localization and dissolve near the Mott transition [4].

References

- [1] M. Dressel and S. Tomić, Adv. Phys. **69**, 1 (2020).
- [2] B. Miksch et al., Science 372, 276 (2021).
- [3] A. Pustogow *et al.*, npj Quantum Materials **6**, 9 (2021); R. Rösslhuber *et al.*, Phys. Rev. B **103**, 125111 (2021); Y. Saito *et al.*, J. Mat. Chem. C **9**, 10841 (2021).
- [4] A. Pustogow et al., Nat. Commun. 12, 1571 (2021).

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