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## (I) Large Kondo Effect in molecule-linked Au Nanoparticles Assemblies

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Interactions between localized, unpaired spins and delocalized electrons play a key role in a range of phenomena, including the Kondo effect, RKKY interactions and high  $T_c$  superconductivity. A number of recent studies have explored such interaction using hybrid systems combining 1) molecules with metal ions which contribute unpaired, localized spins and 2) deposited Au films which contribute delocalized electrons. Such studies have successfully observed a small Kondo effect.

Unexpectedly, a different set of studies have reported that using long (therefore, insulating) alkanethiol ( $\text{CH}_3(\text{CH}_2)_n\text{SH}$ ) molecules as “surface coatings” for Au nanoparticles can generate magnetism in the nanoparticles. Neither bulk gold nor alkanethiols are magnetic by themselves, and it is believed that the thiols ( $-\text{SH}$ ) generate holes in 5d orbitals of gold.

Using short (therefore, conducting) butanedithiol ( $\text{HS}(\text{CH}_2)_4\text{SH}$ ) molecules as crosslinkers for Au nanoparticles, we have observed for the first time a Kondo effect in this molecule linker- Au nanoparticle system. The Kondo effect here is much larger relative to other temperature dependent phenomena –more than 10-fold larger than previously reported in studies using deposited Au films. These results afford testing Kondo models in a much stronger regime, and we find that published theory continues to hold. Importantly, we show that by changing the nanoarchitecture of the films, we can raise the Kondo temperature 10-fold, to  $>250\text{K}$ . These results point to molecule linker-nanoparticle assemblies as a versatile and potentially powerful means to generate materials exhibiting strong electron-electron interactions.

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