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Thema: Redox spin chemistry of transition metal complexes and related donor-complex-acceptor triads

Abstract:

A central aspect of spin chemistry is the explanation and exploitation of magnetic field effects on the kinetics of chemical reactions with radical pair intermediates. The reactivity of such radical pairs is controlled by the electron spin because allowed reactions are restricted by the rule of spin conservation demanding equal multiplicity of radical pair and primary products. Hence, any interaction changing the relative alignment of the two electron spins in a radical pair, will affect its reactive behavior. The most important interactions of this kind comprise hyperfine coupling of the electron spins with the nuclear spins and the Zeeman interaction with an external magnetic field. In metal-centered radicals like Ru(III) and Fe(III) complexes with an open-shell d⁵ configuration, spin-orbit coupling is very strong, which causes their g-factors to strongly deviate from the "free-electron" gₑ of "normal" organic radicals. Thus, in radical pairs composed of such a metal centered radical and an organic radical, the Larmor precession of the two electron spins in an external magnetic field is quite different and leads to fast triplet/singlet transitions, particularly in high magnetic fields. It is shown, how these effects can be exploited to measure very fast electron transfer and spin relaxation processes on the time scale of a few picoseconds.

In a second part of the talk, we will consider electron donor (D) acceptor (A) dyads connected by a [Ru(III)(bpy)₃]²⁺ unit (C), which acts as a sensitizer of charge separation, by which linked radical pairs of type A⁻-C-D⁺ are formed. The life time of these charge separated states exhibits an extremely strong magnetic field dependence, which is quantitatively accounted for in terms of a detailed kinetic scheme involving the specific kinetic behavior of the various spin-substates.

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