# Defect characterisation with coherent spin motion experiments

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Recombination through defects are prominent loss mechanisms of solar cells. The microscopic investigation of these mechanisms is rather difficult since firstly, materials have many recombination paths which are active simultaneously and secondly, recombination can only be observed indirectly through macroscopic observables such as conductivity, capacitance, device performance or luminescense. Hence, any measurement always reveals the superimposed net effect of a variety of recombination paths and electronic processes which makes a straight forward microscopic understanding of recombination difficult if not impossible.

In this study, we present a new method, which we refer to as pulsed electrically detected magnetic resonance (pEDMR), that not only allows the microscopic identification of the recombination mechanism but also gives quantitative access to the dynamics of recombination through paramagnetic defect states. It is based on the recently discovered effect that coherent spin motion of spin pairs that form during the capture of charge carriers at defects can be observed in the macroscopic sample current [1]. The coherent spin motion is induced with pulsed electron spin resonance (ESR) that is tuned into resonance to a specific defect state. The selectivity is obtained through the fact that the Landé g-factor of the defect depends on the microscopic environment. Under illumination, a constant photocurrent and thus a steady state recombination rate is obtained as shown in Fig. 1a. Spin-pairs are always generated at random in the four different pair eigenstates. Since the lifetime of triplet pairs is significantly longer than that of singlets, a surplus of triplets will prevail under steady state conditions, as indicated by the two parallel spins in Fig. 1a. If a microwave that is in resonance with either of the two spin-pair partners (e.g. electron or defect) is switched on (Fig. 1b), a relative spin motion between the two spin partners will start (Rabi oscillation) that induces an oscillation of the singlet content of the spin pairs and hence of the capture and recombination rate. This may then be reflected as a small oscillation of the photocurrent. The coherence decay of this oscillation is determined by the singlet capture cross section of the defect providing that no other, faster source of decoherence exists. After the microwave burst is turned off, fast Lamor oscillations take place and thereafter the photocurrent slowly relaxes back to its steady state (Fig. 1c-e). The shape of this relaxation transient is determined

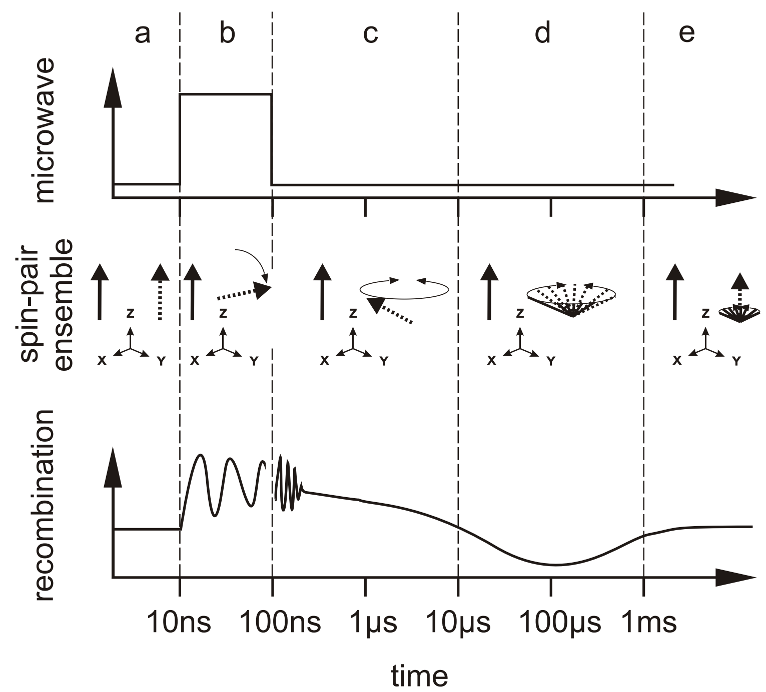


**Figure 1:** Sketch of the time evolution of the spin-dependent recombination rate of charge carriers at paramagnetic defects before, during and after a microwave burst. Note that the illustration does not reflect any experimental or simulated data and is intended only to visualise the different processes that take place.

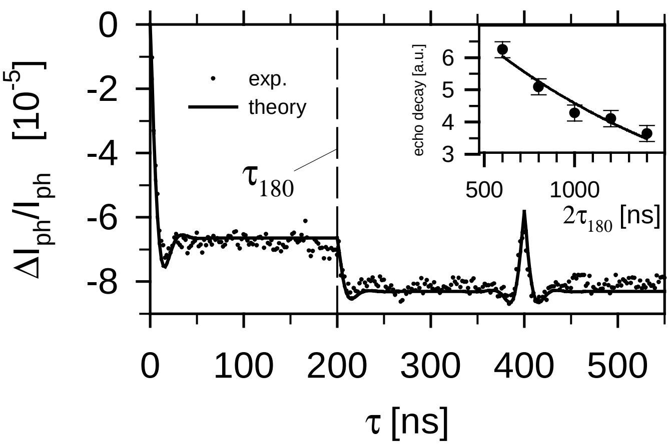


**Figure 2:** Experimental data (dots) of the photocurrent transient during the application of a strong microwave pulse in resonance with dbs in µc-Si:H recorded at T = 10 K. The phase change is introduced at $\tau_{180}$ = 200 ns leading to an echo in the photocurrent at 400 ns. The solid line represents a calculated transient assuming strong spin-spin coupling [1]. The inset shows the coherence decay of the echo intensity.

by slower incoherent processes such as triplet recombination as well as by spin relaxation and pair dissociation [2]. PEDMR is not only a highly selective method, it is also highly sensitive. For state of the art current detection technology, it is possible to detect as little as 100 recombination events in a given sample. This is about $10^{10}$ times better than conventional electron spin resonance.

The feasibility of pEDMR is demonstrated on dangling bond (db) recombination in hydrogenated microcrystalline silicon (µc-Si:H). The photoconductivity response during the application of a microwave burst that is in resonance with the db is shown in Fig. 2. A current oscillation (and hence the direct observation of the coherence decay) cannot be observed since inhomogeneously distributed Rabi frequencies lead to a strong (coherent) damping of the current oscillation. However, by changing the phase of the microwave by 180° at $\tau_{180}$ = 200 ns, the coherent damping can be rephased leading to an echo of the photocurrent at $\tau$ = 400 ns. The step-like feature at the moment of the phase change is an inherent property of the recombination mechanism. The solid line in Fig. 2 is a calculation of the photocurrent response assuming that the recombination mechanism is determined by direct capture (dc) of charge carriers into charged, excited $db^{-*}$ states with energies close to the conduction band, from which realignment transitions into the charged $db^{-}$ ground states occur. Recombination is completed by hole capture. Such a process is, compared to a distant pair recombination, determined by strong spin-spin coupling between the charge carriers and the db. Note that the excited $db^{-*}$ states in µc-Si:H cannot be observed by conventional defect spectroscopy due to the high density of tail states present in the material. The simulation nicely reproduces all the features of the experiment, even the fact that the echo intensity does not fully recover although no incoherence is assumed in the calculation.

By measuring the echo intensity as a function of $\tau_{180}$ we are able to determine the coherence decay of the spin-pair ensemble that is located at the dbs. As shown in the inset of Fig. 2, the decay can be fitted by a single exponential leading to a coherence decay of 1.2(3)µs. Since the spin relaxation times $T_1$ and $T_2$ are much smaller than this value at the given temperature, we can identify this time with the recombination time from singlet states. As was shown before [3] we find that the triplet state of the spin pair which causes the observed signal has a lifetime that is about a factor of 100 lager then that of the singlet state. Since the singlet and triplet lifetimes are independent on the g value of the db, we conclude that the different microscopic environments of the db seem to have no influence on the capture cross section of dbs. This is further evidence for the correctness of the $db^{-*}$ recombination model.

References

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[3] C. Boehme, K. Lips, Mat. Res. Soc. Symp. Proc. **715**, A16.2 (2002).