Auto-correlation. An initial analysis of the distribution of the slow-on and slow-off states' survival times showed an exponential decay with time, indicating a stochastic process[1] . However, the lifetimes of the fast-on and fast-off states were examined using the higher time-resolution technique (higher even than the sampling rate) of auto-correlation[2] of the time-resolved current measurements *I(t)*,

$$g^{(2)}(\delta) = \langle I(t)I(t+\delta)\rangle \big/ \langle I(t)\rangle^2 .$$

Knowing that the distribution of survival times for both states is exponential, the auto-correlation was modelled by,

$$g^{(2)}(\delta) = 1 + C\exp\left[-\left(1/\tau_{On} + 1/\tau_{Off}\right)\delta\right],$$

with τ_{On} and τ_{Off} the lifetimes of the on- and off-states.[2]

With well defined on- and off-states the contrast $C = \left(I_{On} - I_{Off}\right)^2 \Big/ \left[\tau_{On}\tau_{Off}\left(I_{On}/\tau_{On} + I_{Off}/\tau_{Off}\right)\right]$ with I_{On} and I_{Off} the tunnelling current values determined from the Gaussian fits to the current distributions. Within experimental error, both the auto-correlation and the distribution of survival time analysis gave the same lifetime values for the on-state and off-states for the slow-switch.

When only one state was well defined the other state's tunnelling current value, for example, I_{On} was substituted by $\left(\tau_{Off} I_{Off} + I_{Average}\tau_{On}\tau_{Off}\right)\big/\tau_{On}$ where $I_{Average}$ is the average tunnelling current over the whole time-trace. From the resulting lifetime values the unknown tunnelling current was calculated.

References

[1] Lastapis, M.; Martin, M.; Riedel, D.; Hellner, L.; Comtet, G.; Dujardin, G. *Science* **2005**, *308*, 1000.

[2] Zumbusch, A.; Fleury, L.; Brown, R.; Bernard, J.; Orrit, M. *Phys. Rev. Lett*. **1993**, *70*, 3584.