Ultrafast Processes in Functionalized Geramanane

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Methyl-substituted germanane $GeCH_3$ is an emerging layered two-dimensional semiconductor proposed for novel applications in optoelectronics, photoelectrocatalysis, and biosensors. Its photoluminescence (PL) spectrum can be reversibly switched between a bright red peak at 1.97 eV upon water intercalation, and a broad band-tail emission for the dry material [1].

The time-resolved photoluminescence dynamics of a $GeCH_3$ crystal with a moderate amount of water intercalation originates from exciton rather than free carrier recombination [2]. Remarkably, the dynamics depend nonmonotonically on temperature as it becomes gradually slower with increasing temperature, while at higher temperatures they quickly become faster again. We identify two exciton populations, which can both be excited and relaxed towards the ground state with weakly temperature-dependent lifetimes. Additionally, a thermally activated transition from the shorter to the longer-lived species is observed.

To gain a deeper insight into the early processes after photoexcitation, we studied the TA in a pump-probe configuration. We used as the sample a dispersion of $GeCH_3$ flakes in anhydrous isopropanol obtained via liquid phase exfoliation [3].

The TA results point towards a more complex relaxation cascade that starts with hot carrier cooling on a sub-ps timescale that is slower for higher fluences, consistent with a phonon bottleneck.

In conclusion, time-resolved PL and TA investigations unveiled a rich photoexcitation dynamic in $GeCH_3$ interacting with polar molecules. The main photoexcited species are two distinct exciton populations that couple primarily to different phonon modes.

References

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