Ultrafast Nanorotor

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Introduction

The optically active nanoparticles and dispersion have shown promising application in the field of energy harvesting, photo-thermal, photonics, and sensing. The gyration in the photo-excited charge dynamics have a great interest for practical application of the anisotropic nanoparticles (e.g. nanorods (NRs) dispersion). As per the electric field-dipole approximation ($\vec{\mu}.\vec{E} \propto \cos^2\theta$), the orientation between the photo-excited dipoles, and emitted dipoles may change due to the rotation of the dispersed NRs under the influence of electromagnetic field within the time-frame of lifetime of the dipole. The excited states of the NRs, where the carrier exhibits longitudinal and transverse modes of localization, acquit anisotropic emission depending upon the orientation of the crystal. Thus, the optical properties of the NRs dispersed in a fluid, where they experience gyration under an external electromagnetic field is an important point of debate for application in the nanomechanical devices.

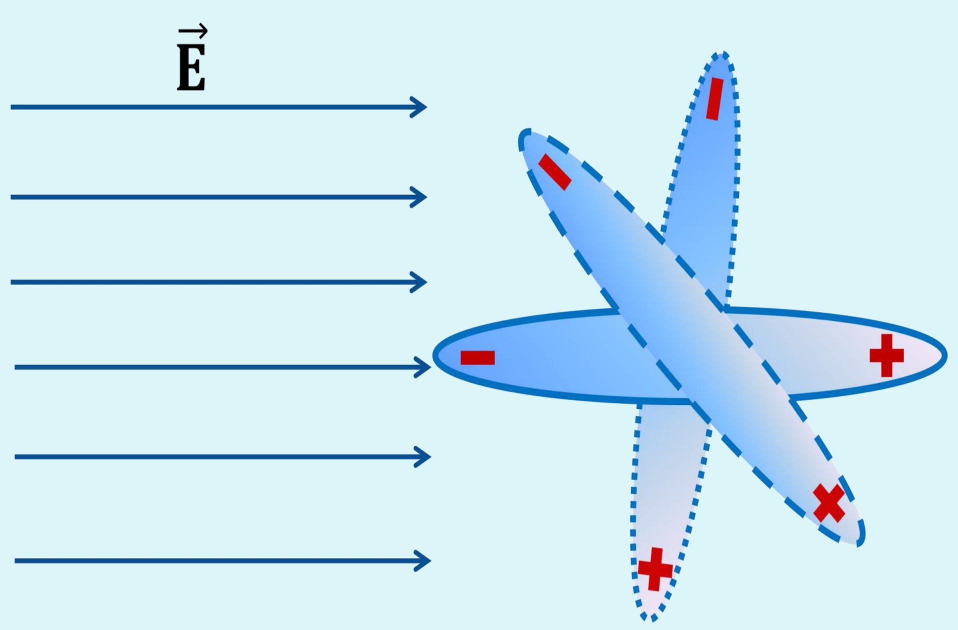
Experimental Method

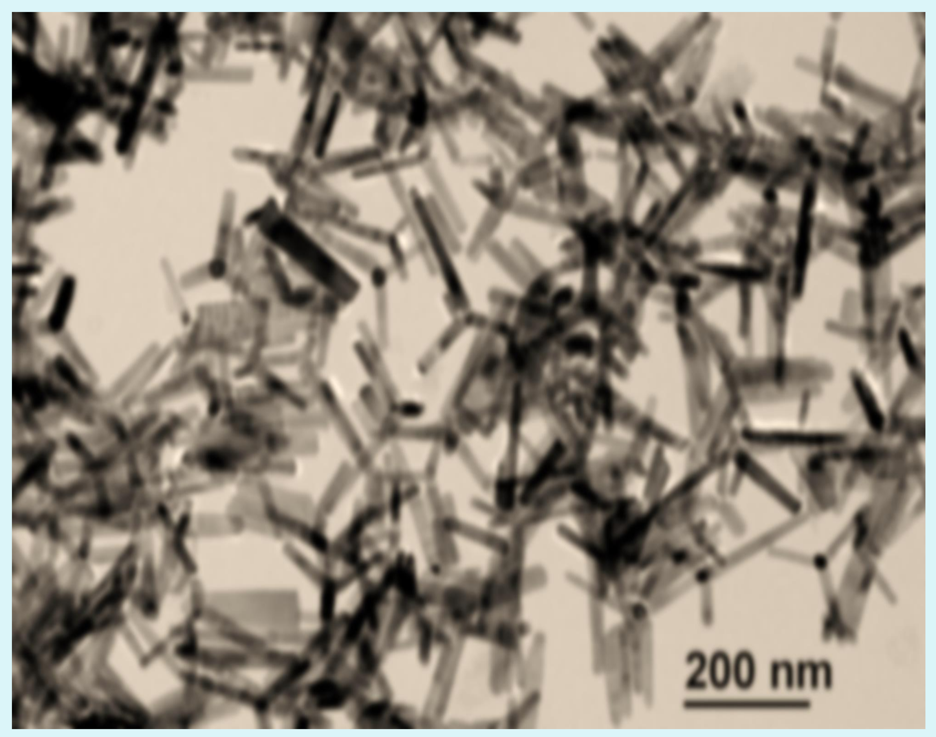
The CdS nanopods with aspect ratio of 5.30 have been synthesized using a micellar growth technique. The extracted CdS nanorod powders were dispersed in the chloroform for the PTRES and TDA measurements.

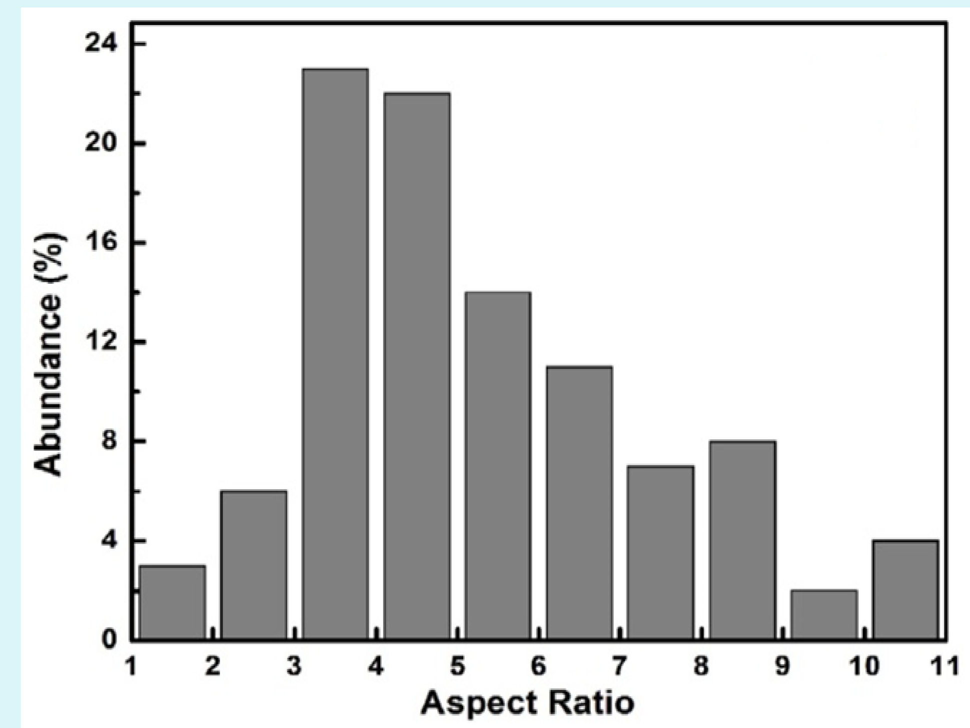
Objectives

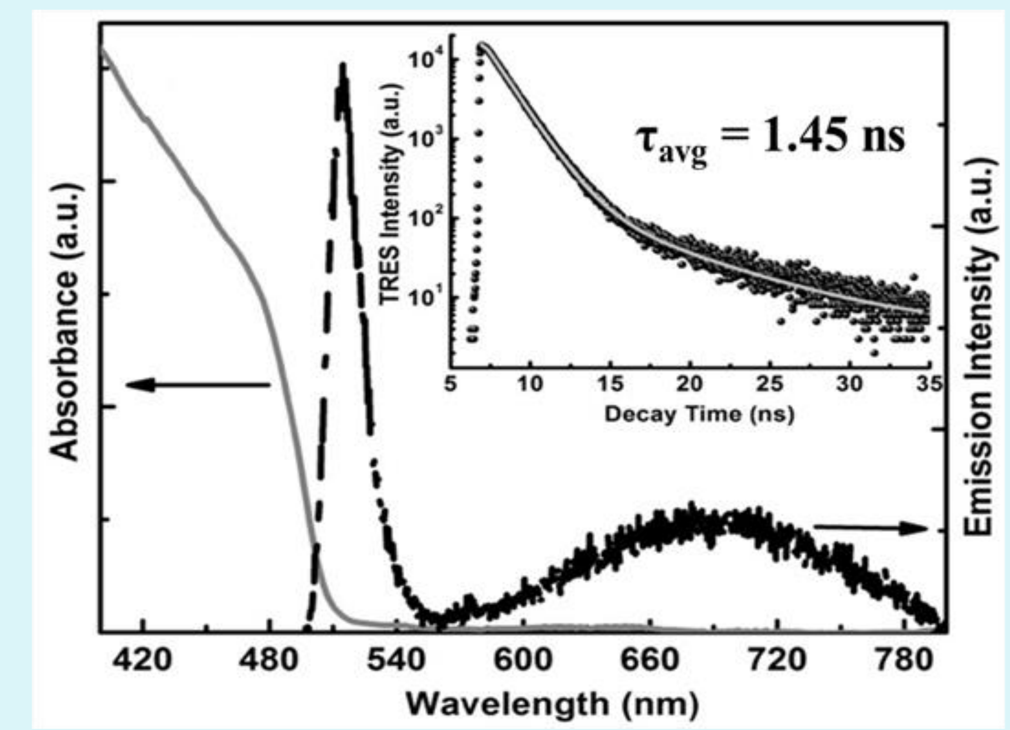
- To study the rotational dynamics of nanopods as a nanomechanical rotor.
- The role of excited dipoles in the ultrafast rotation.
- Calculation of the time period of rotation with the help of Time dependent Anisotropic analysis through Time resolved photoluminescence spectroscopy.

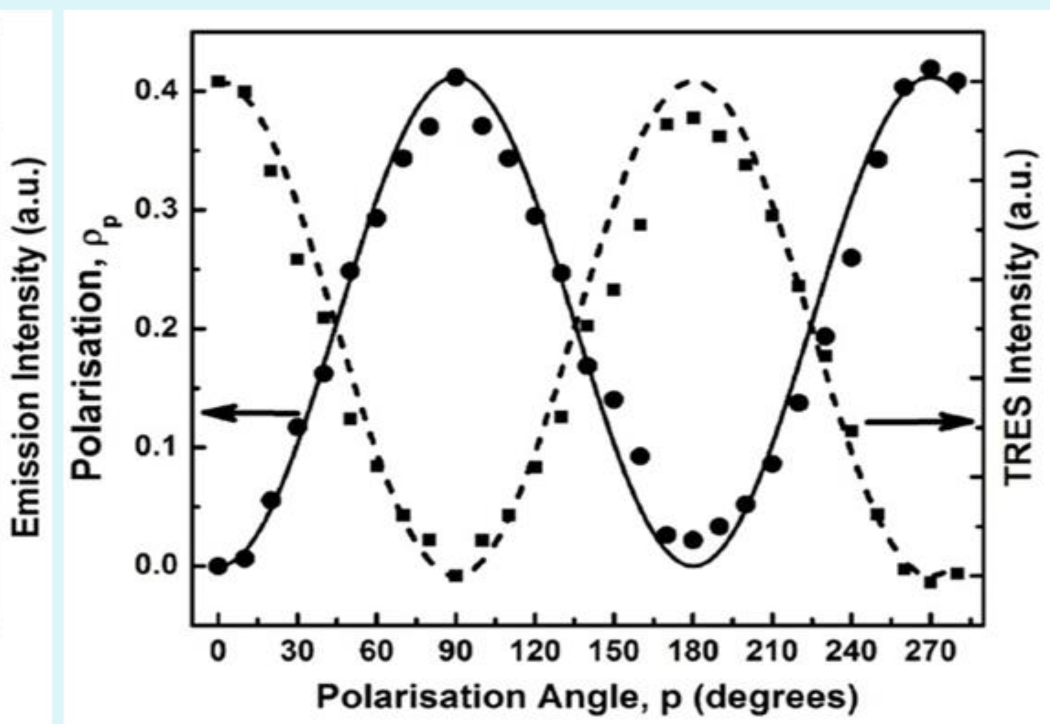
Results & Discussion



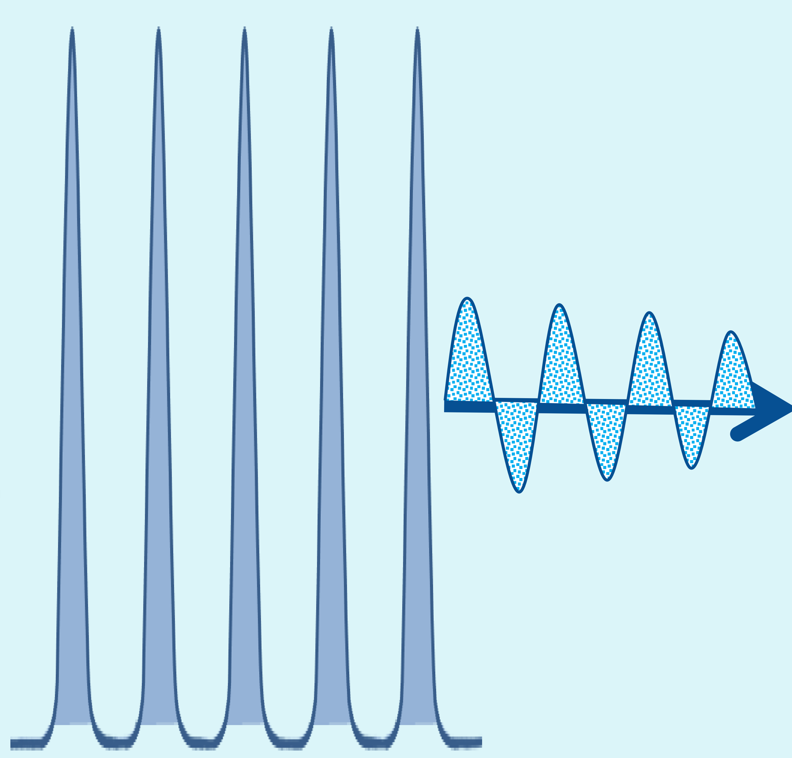


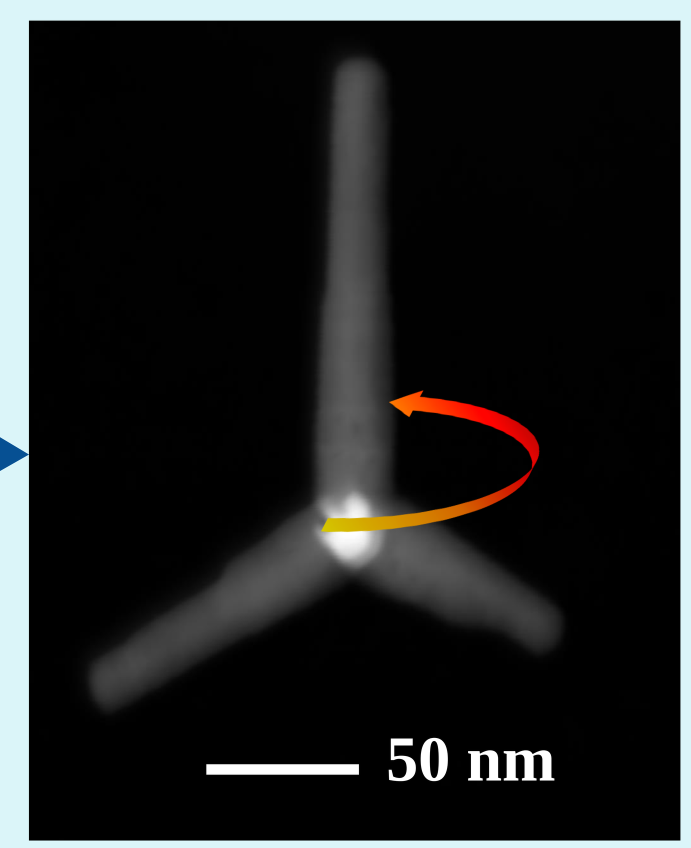


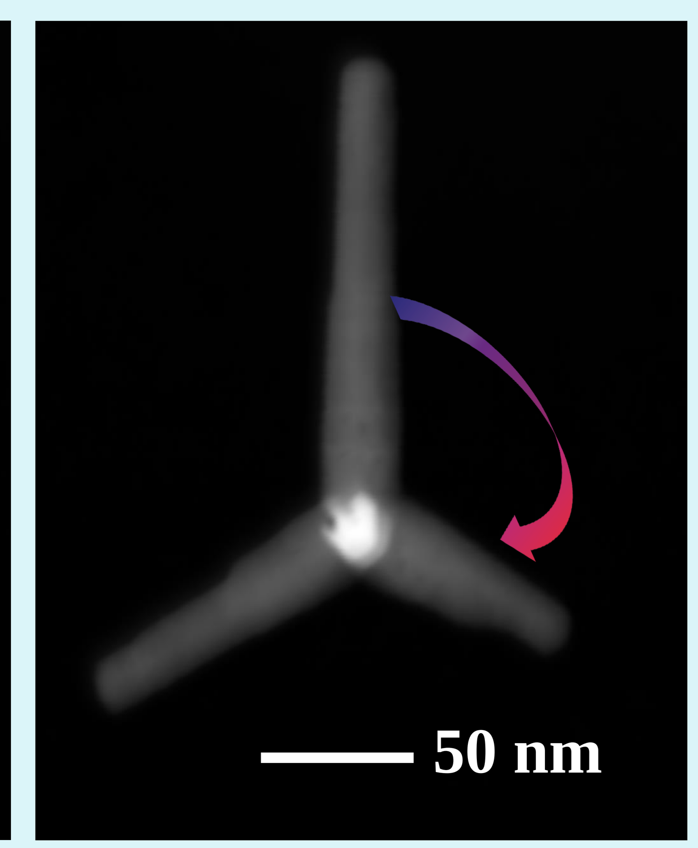




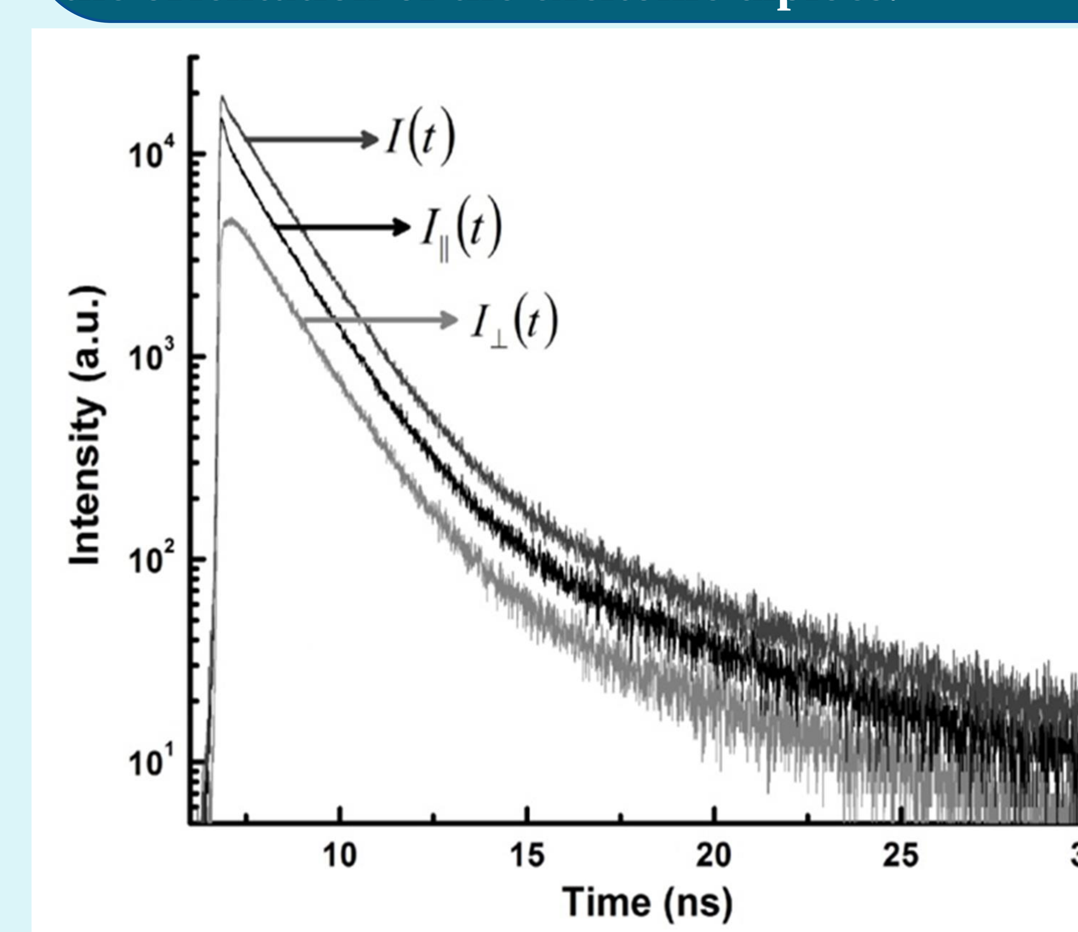
- TEM image confirms, the sample exhibits a distribution of nanorods joined at a common end forming nanopod structure with aspect ratio 5.30.
- Two possible ways of rotation occures (e.g. perpendicular and parallel axis of rotation) in the presence of pulsed diode laser (Excitation wavelength 375 nm and 1 MHz pulse rate).

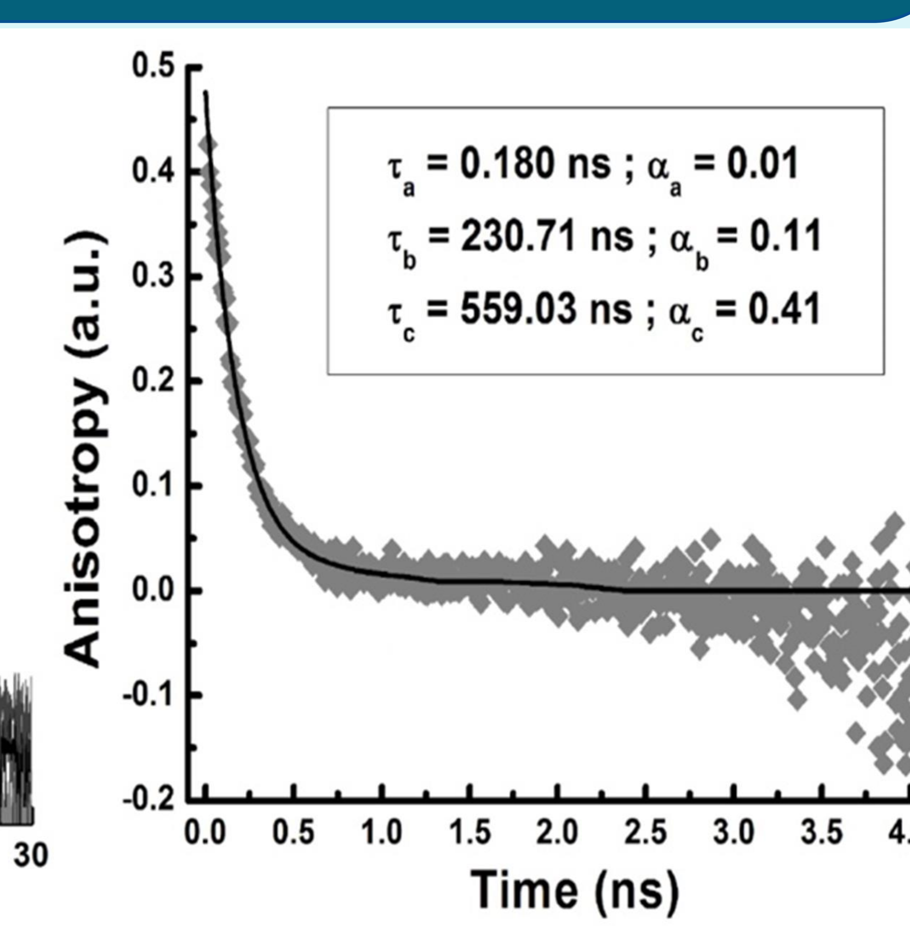






By using the double exponential decay fitting in the time resolved emission spectra, the average life time of the exciton $[<\tau>=\frac{\alpha_1\tau_1^2+\alpha_2\tau_2^2}{\alpha_1\tau_1+\alpha_2\tau_2}]$ is calculated as 1.45 ns. In order to understand the effect of electromagnetic field induced rotation, the TRES measurement has been carried out at different polarization angle. The degree of polarization has been evaluated using the relation, $\rho_p=\frac{I_{\parallel}-I_p}{I_{\parallel}+I_p}$, as 42%, which is corresponding to a change in the orientation of the excitonic diploes.





The shape anisotropy on the dynamics of the excited state has been investigated using the TDA measurements. The integral decay intensity, $I(t)=I_{\parallel}(t)+2I_{\perp}(t)$ and anisotropic decay, $r(t)=\frac{I_{\parallel}(t)-I_{\perp}(t)}{I(t)}$ are shown in the above figures. The uncorrelated emission lifetimes and rotational correlation times are related through the anisotropic decay, $r(t)=\alpha_a e^{\frac{-t}{\tau_a}}+\alpha_b e^{\frac{-t}{\tau_b}}+\alpha_c e^{\frac{-t}{\tau_c}}$. By using the triple exponential decay fitting the time period of rotation around the parallel and perpendicular axis are estimated to be 230.71 ns and 559.03 ns respectively.

Summary

- The co-related optical and mechanical properties of the CdS nanopods with an aspect ratio as 5.30 have been discussed.
- The optical properties of the nanopods dispersed in the Chloroform show a strong band edge emission at wavelength 514 nm.
- The excited states dynamics of the sample exhibits the polarization anisotropy as 42 % due to the dipole-dipole interaction mediated ultrafast rotation.
- From the time dependent anisotropic studies, the nanopods observed to be exhibiting rotational time period ~ 230.71 ns and 559.03 ns along the parallel and perpendicular axis of rotation respectively.

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