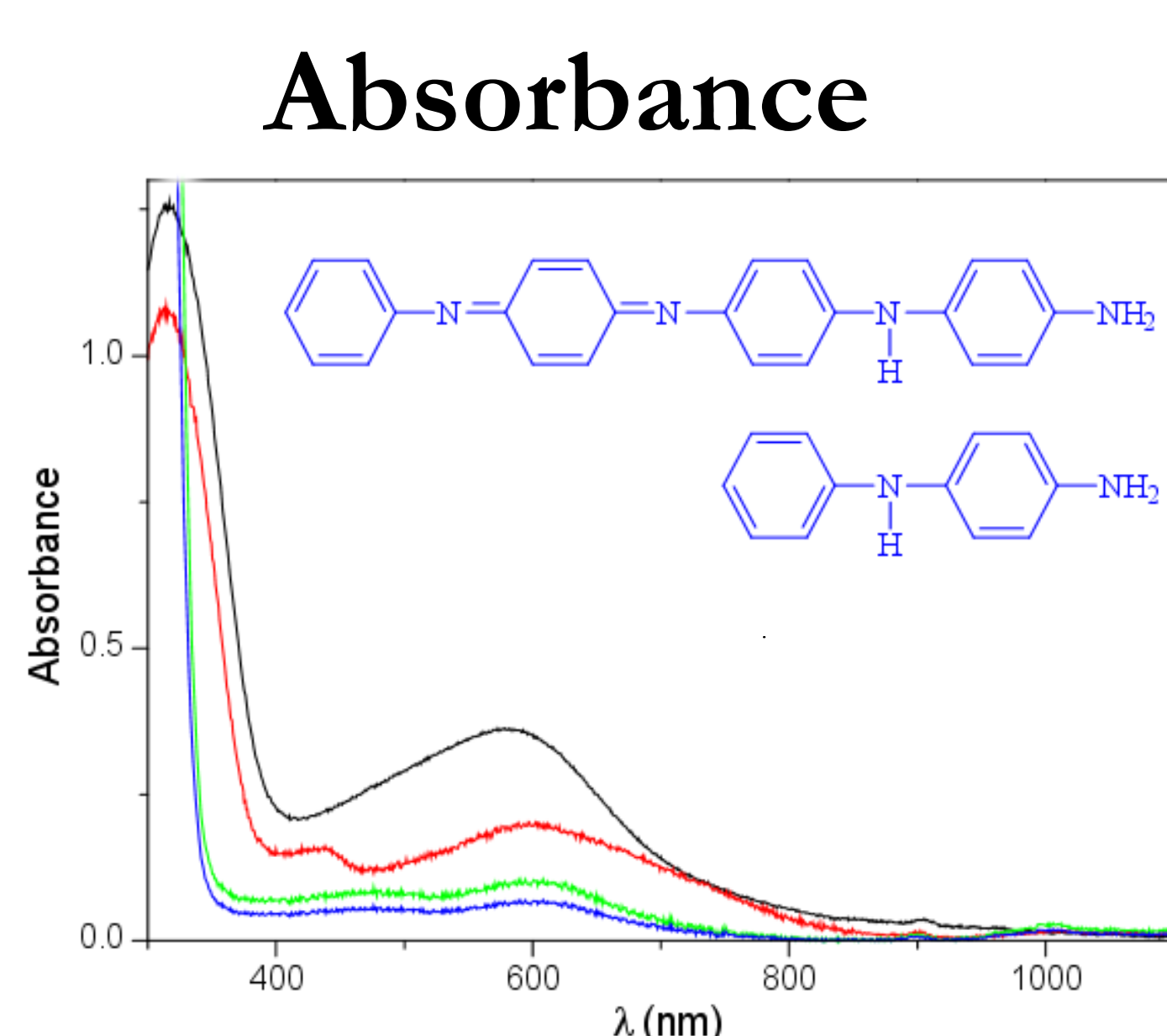
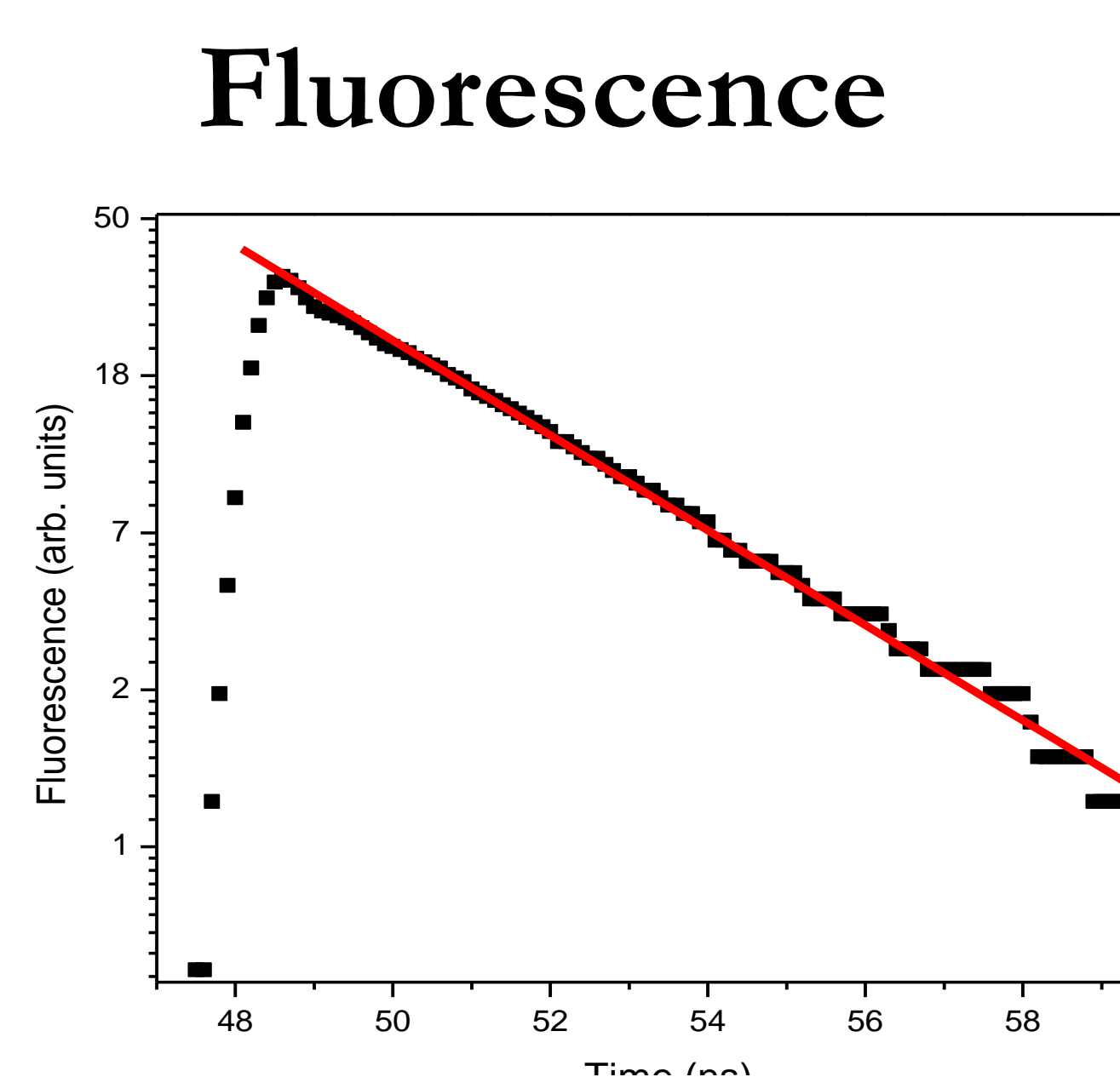


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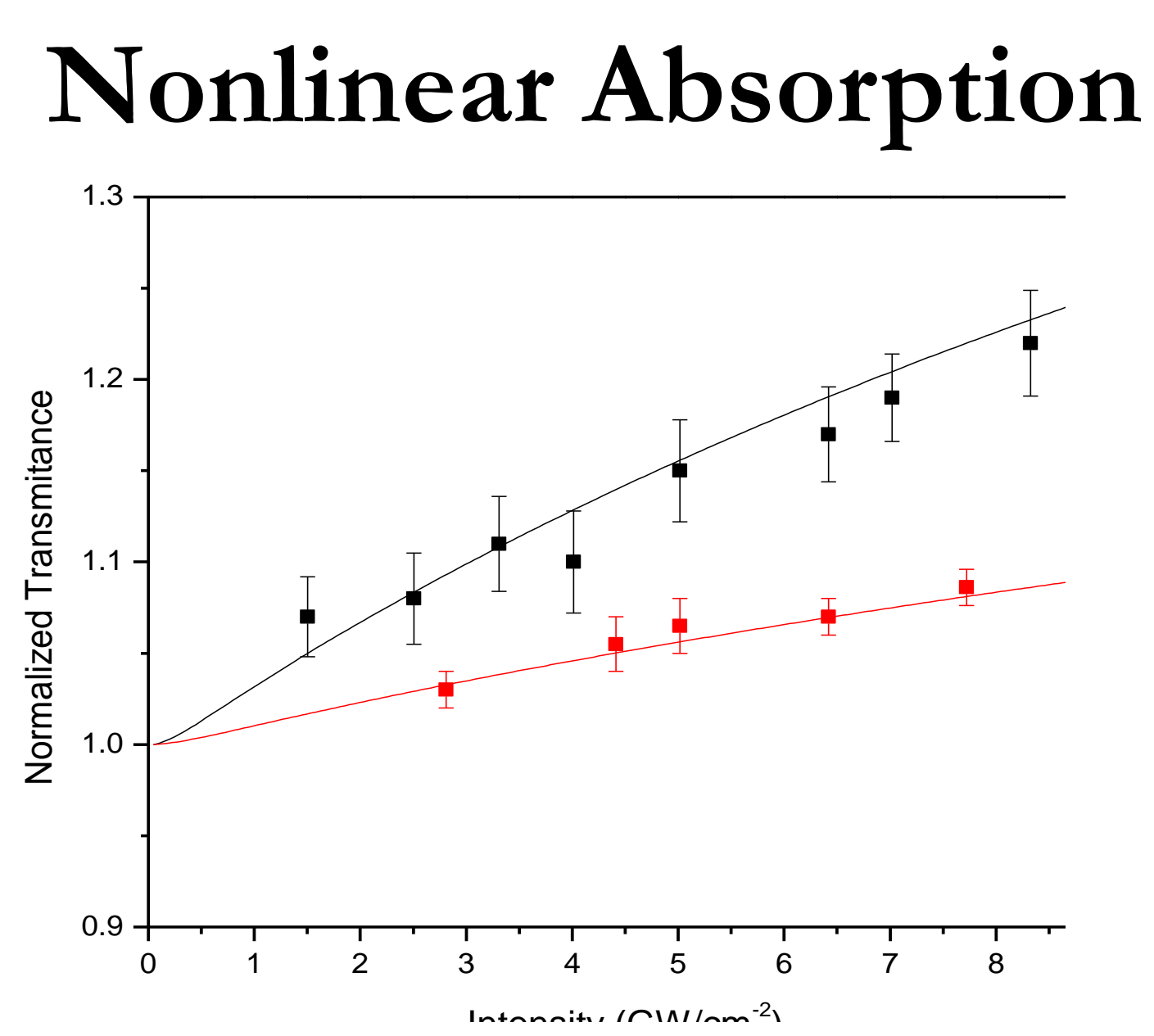
In this work, we investigate the nonlinear optical properties of the phenyl/ NH_2 end capped dianiline and tetraaniline molecules. We employed the techniques of linear spectroscopy, time-resolved-fluorescence, Z-scan and Hyper-Rayleigh scattering to perform the optical measurements. A three level model was proposed to explain the electronic mechanism responsible for the saturated absorption. Our results shows that the doping process modestly diminishes the nonlinearity at probe wavelength (532 nm), and the magnitude of nonlinearities is comparable with much longer chain length polyanilines. We conclude that the usually verified dependence of nonlinearity on π -electron mobility and chain length no longer holds for organic absorbers.



Linear Absorption and structure of tetraaniline and dianiline.



Fluorescence decay time for a 532 nm excitation .



Nonlinear absorption of undoped (black) and doped (red) tetraaniline.

Rate equations

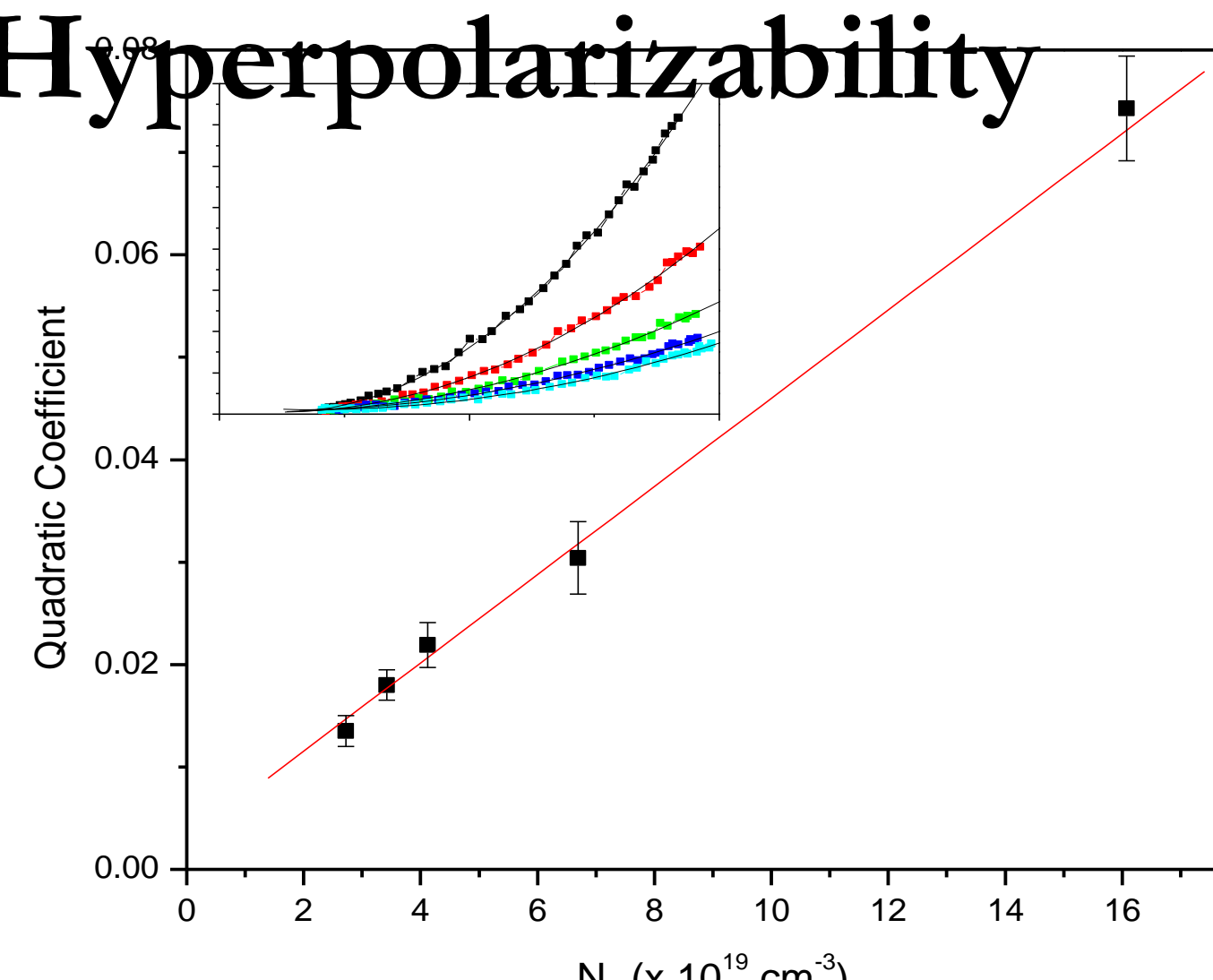
$$\frac{dn_0}{dt} = -n_0 W_{01} + \frac{n_1}{\tau_{01}}$$

$$\frac{dn_1}{dt} = n_0 W_{01} - n_1 W_{12} + \frac{n_2}{\tau_{21}} - \frac{n_1}{\tau_{10}}$$

$$\alpha(I) = N(n_0 \sigma_{01} + n_1 \sigma_{12})$$

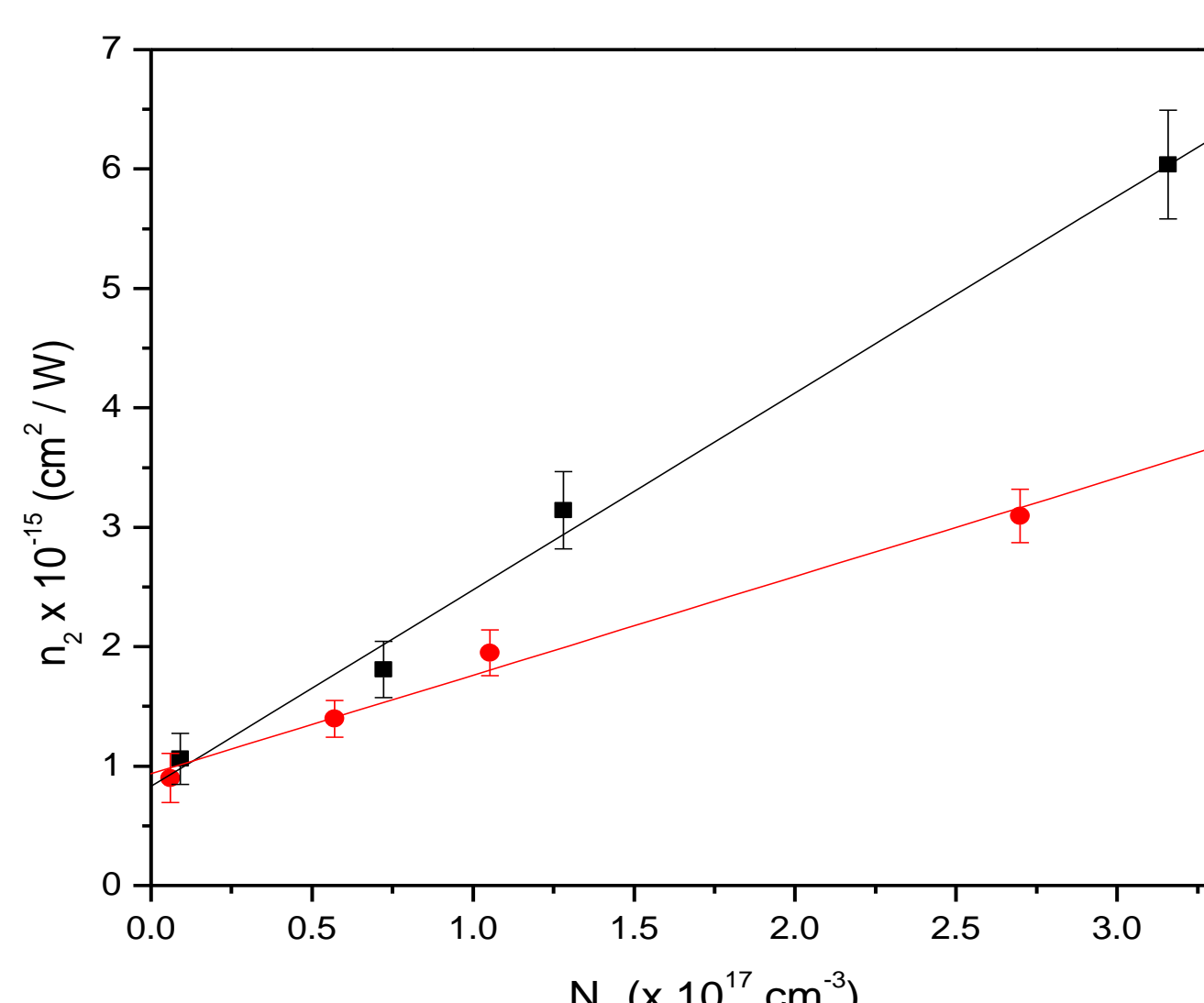
$$W_{01} = \sigma_{12} I_0 / \hbar \omega$$

First Hyperpolarizability

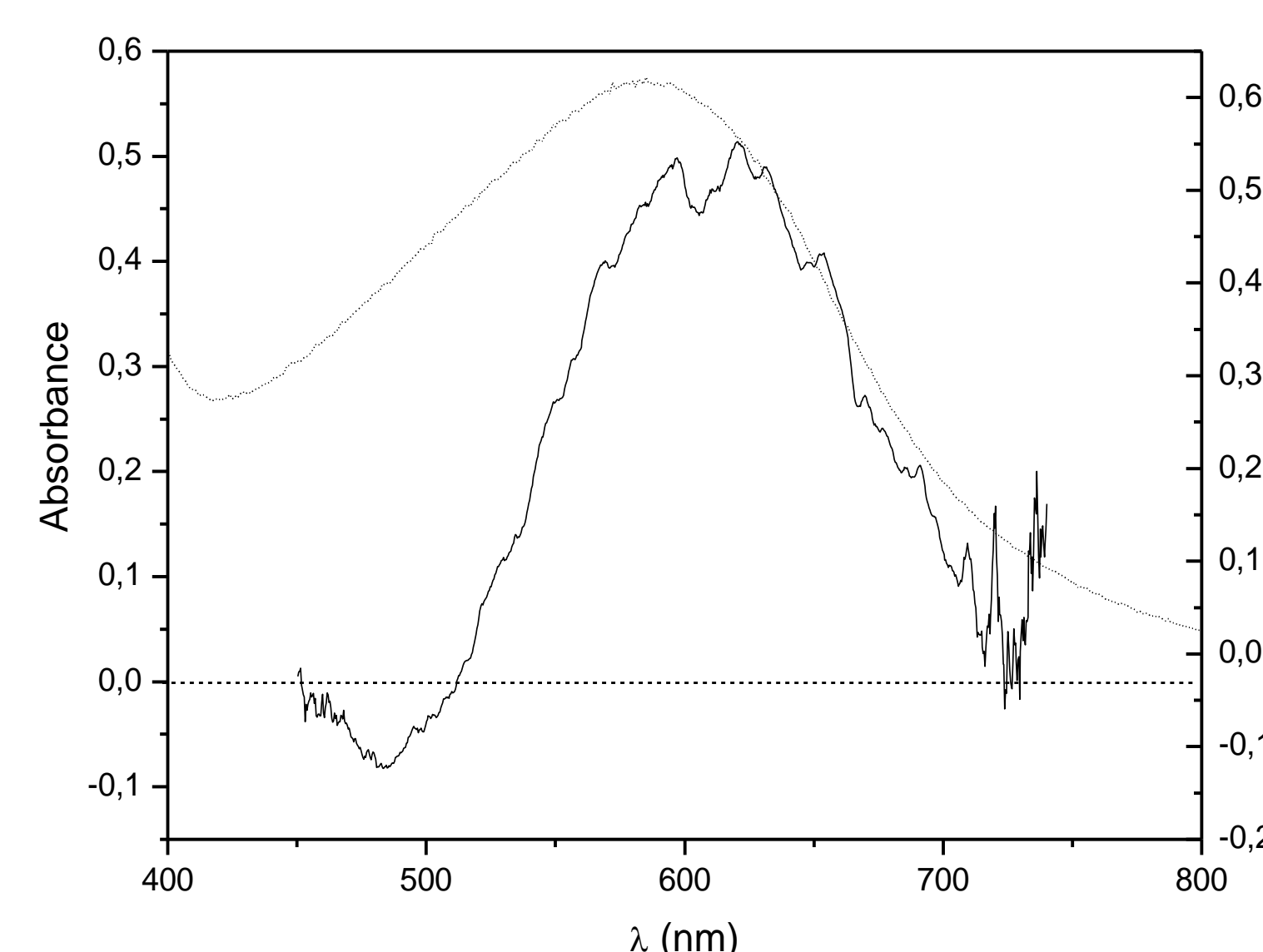


Results of HRS measurement on undoped tetraaniline.

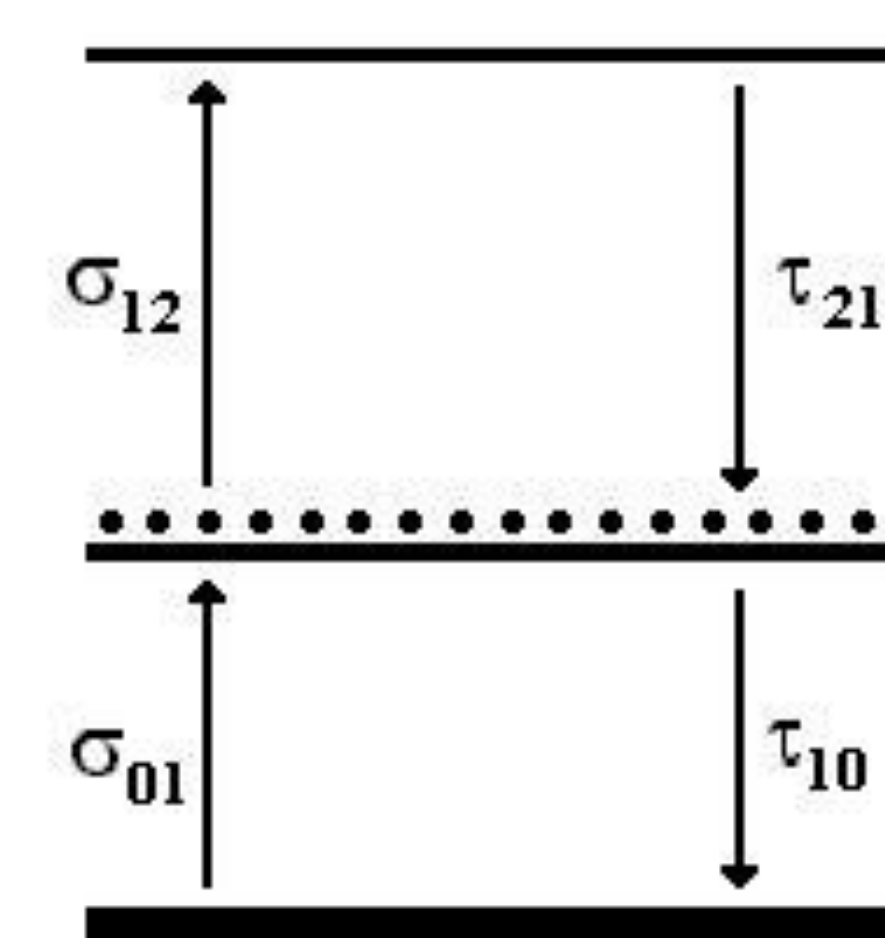
Nonlinear Refraction



Second order refraction as measured for the undoped (black) and doped (red) tetraaniline.



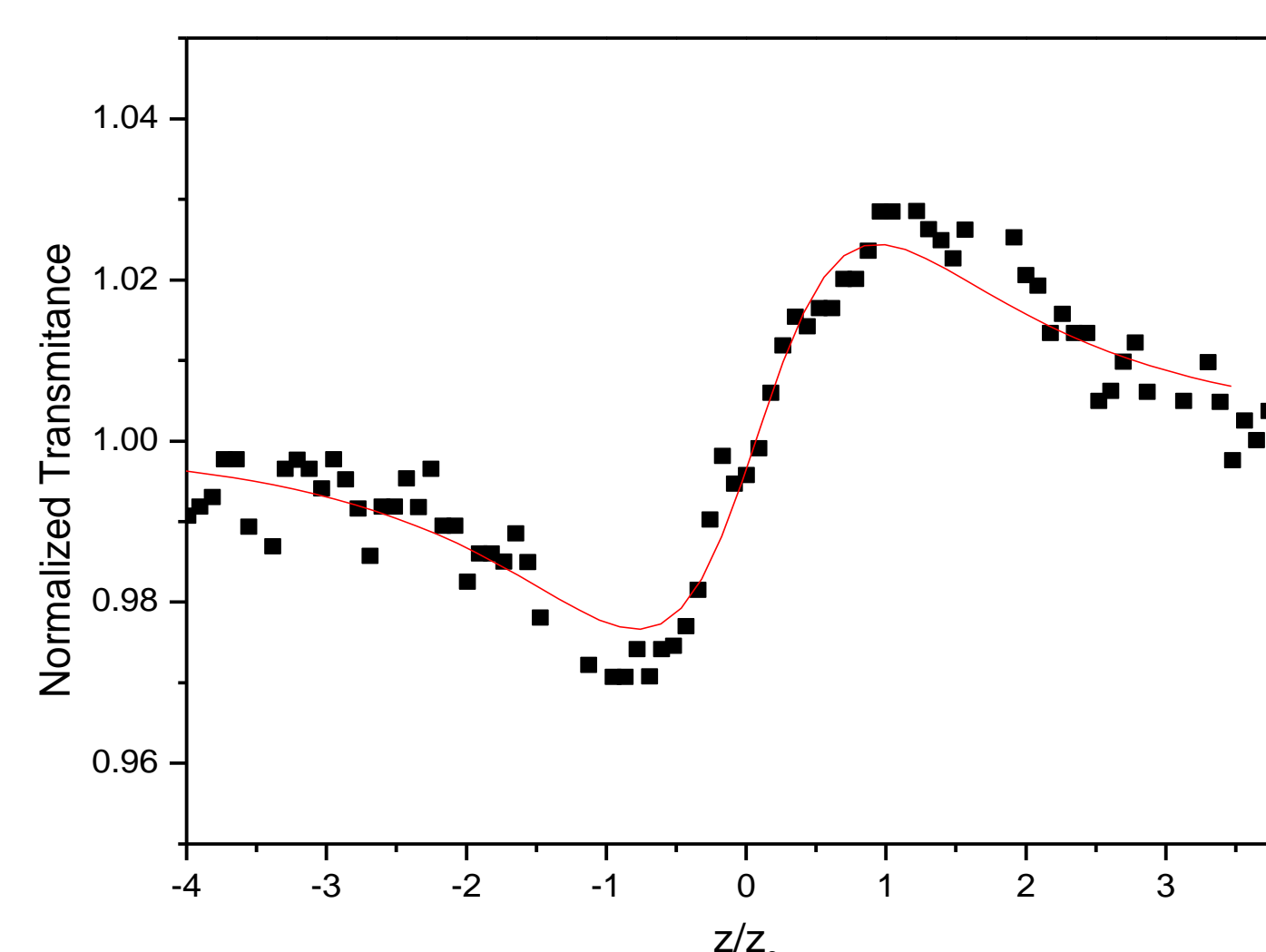
Comparison between the linear and non-linear absorption behavior.



The three-level diagram used for numerical simulation of absorption data.

First hyperpolarizability (β) (in units of 10^{-30} esu)

Molecule	Undoped	Doped
Dianiline	95.6	182
Tetraaniline	645	1475



Z-scan signature and numerical simulation for tetraaniline.

Conclusions

Our results show that the doping process modestly diminishes the nonlinearity at probe wavelength (532 nm), and can be explained by decrease in the linear absorption coefficient. In counterpart, the magnitude of nonlinearities is comparable with much longer chain length polyanilines. Therefore, we conclude that the usually verified dependence of nonlinearity on π -electron mobility and chain length no longer holds for organic absorbers.