Non-linear response of low dimensional structures

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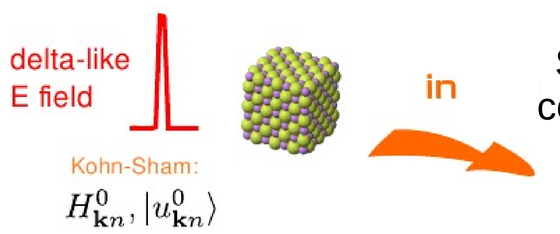
We present an *ab-initio* study of non-linear response of low dimensional structures. In particular we investigate second-harmonic generation (SHG) in two dimensional crystals, MoS_2, h-BN, ZnO, GaN and SiC, and third-harmonic-generation (THG) in carbon nanotubes and nanoribbons. The non-linear response functions are obtained from a real-time approach based on dynamical Berry phase to threat the coupling between the external field and electrons.

Theory: a real-time approach to study non-linear response functions

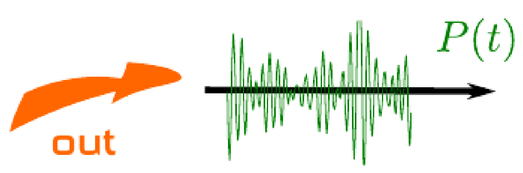


We start from the Kohn-Sham eigenvectors and eigenvalues plus an external electric field

We propagate in real-time a Schroedinger like equation where correlation effects are included as a single particle operators in $H^0_{\mathbf{k}}$

Solve Euler-Lagrange equations:

$$i|\dot{v}_{\mathbf{k},m}\rangle = \left(\hat{H}^0_{\mathbf{k}} + \hat{w}_{\mathbf{k}}(\boldsymbol{\mathcal{E}}) + \hat{w}^{\dagger}_{\mathbf{k}}(\boldsymbol{\mathcal{E}})\right)|v_{\mathbf{k},m}\rangle$$



Polarization is calculated in terms of dynamical Berry-Phase
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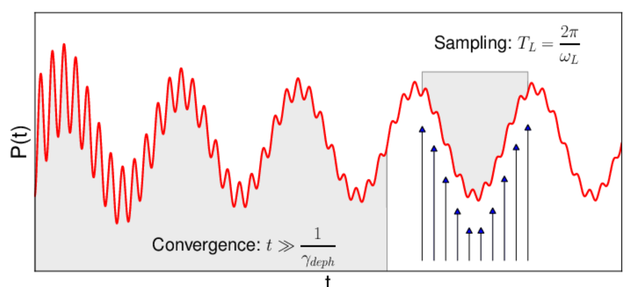
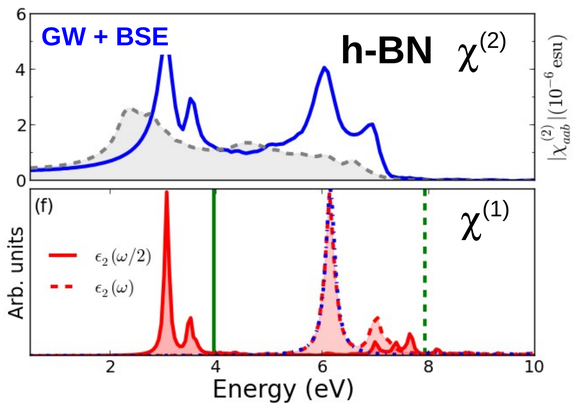
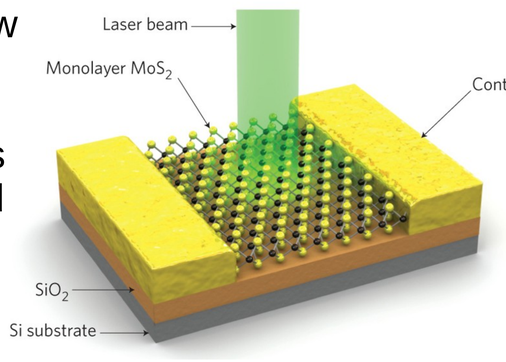


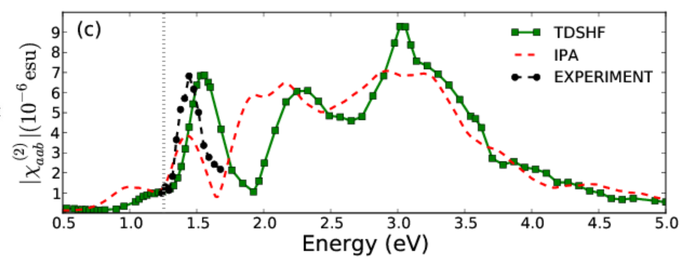
Fourier analysis is used to extract all the response functions $\chi^{(1)}$,$\chi^{(2)}$...

Second Harmonic Generation (SHG) in 2D crystals

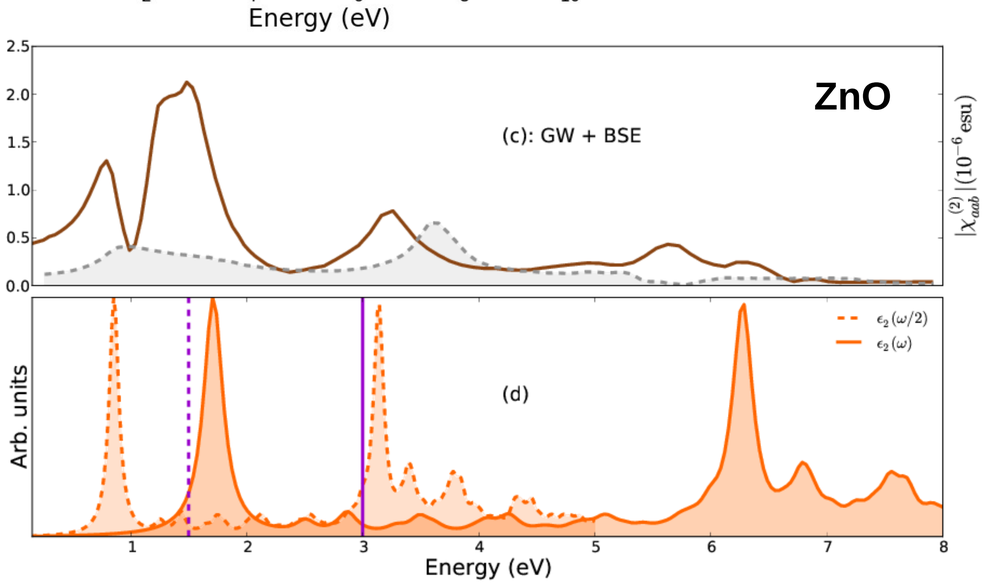


Correlation effects are larger in low dimensional systems. In this example we show the SHG of an **h-BN monolayer**. Bound excitons are clearly visible in the linear and non-linear response
PRB **89**, 081102(R) (2014)

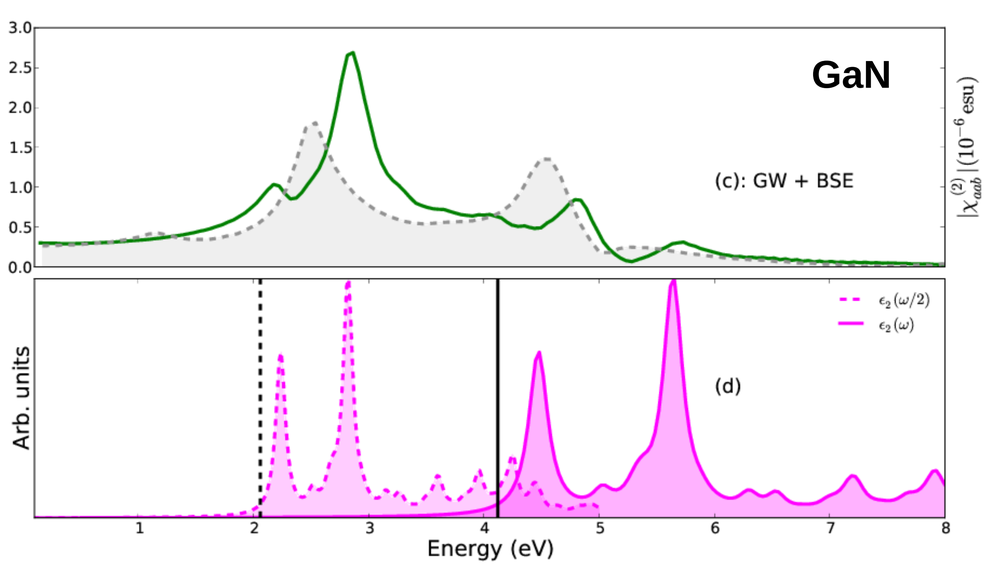




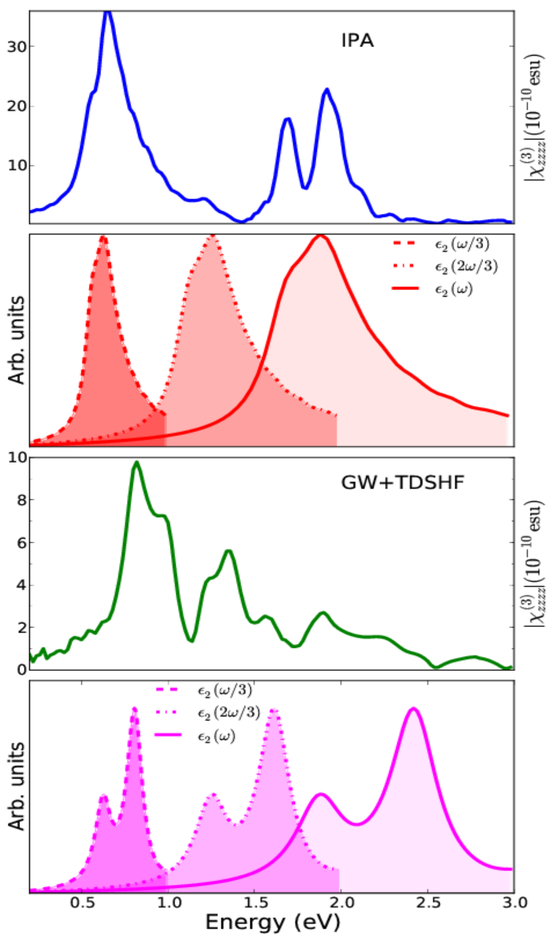
SHG in MoS_2 comparison with experiments from PRB **87**, 201401(R) (2013), the electron-hole interaction enhances the SHG



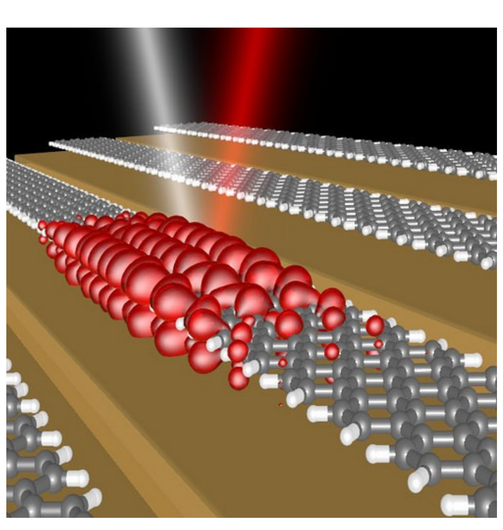
SHG in two-dimensional Hexagonal ZnO and GaN

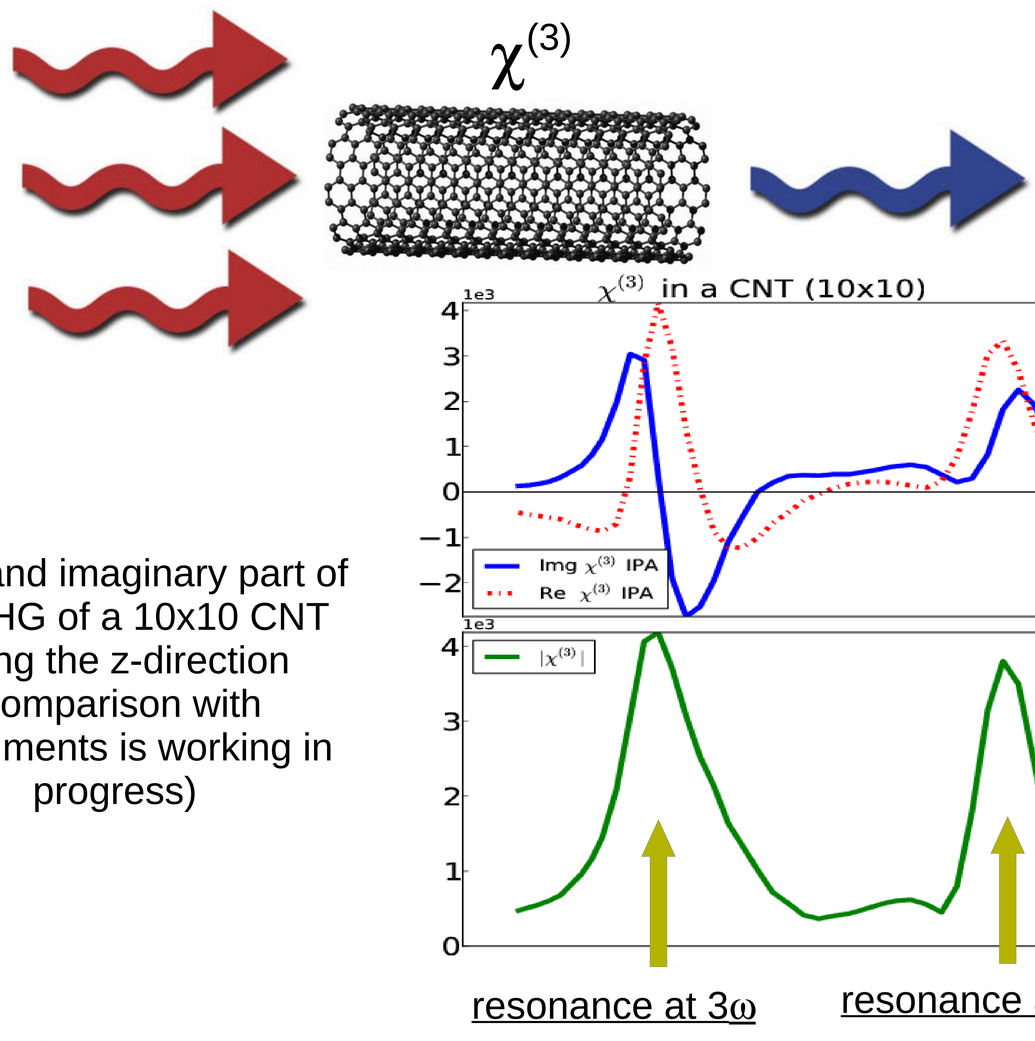


Third-Harmonic Generation in nanotubes and nanoribbons



Third-Harmonic Generation in a carbon nanoribon, compared with the imaginary part of the dielectric constant calculated at ω and 3ω





Real and imaginary part of the THG of a 10x10 CNT along the z-direction (comparison with experiments is working in progress)

resonance at 3ω

resonance at ω

Conclusions

We studied non-linear response of different nanostructure by means of a novel and efficient approach in real-time. We showed that excitons play a major role not only in the linear response but also in the non-linear one due to the reduce dimensionality and the poor screening.