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FIRST-PRINCIPLES HIGH-THROUGHPUT APPROACH FOR LINEAR AND NONLINEAR OPTICAL PROPERTIES

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NOVEL LINEAR AND NONLINEAR OPTICAL MATERIALS

Exploration and prediction via *ab-initio* methods of novel materials with nonlinear properties

Optical application:

- Communications
- Energy
- Security
- Health and Medicine

Nonlinear optical materials: A common application

Green light (532 nm) in laser pointer is obtained via a nonlinear crystal (KTP)

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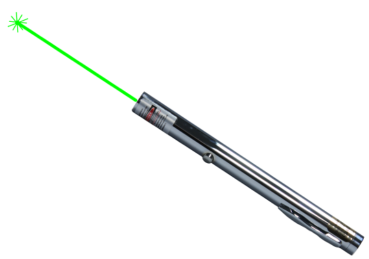
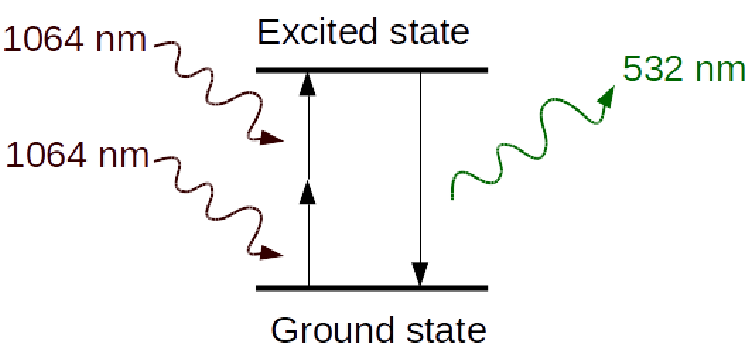
SECOND HARMONIC GENERATION (SHG)



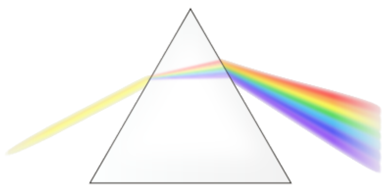


Annu. Rev. Mater. Res., 45, 491(2015);

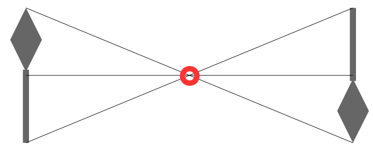
SECOND HARMONIC GENERATION CRYSTALS

Which properties are required to have a “good” material?

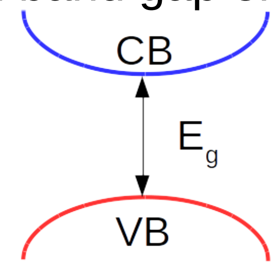
Large refractive index

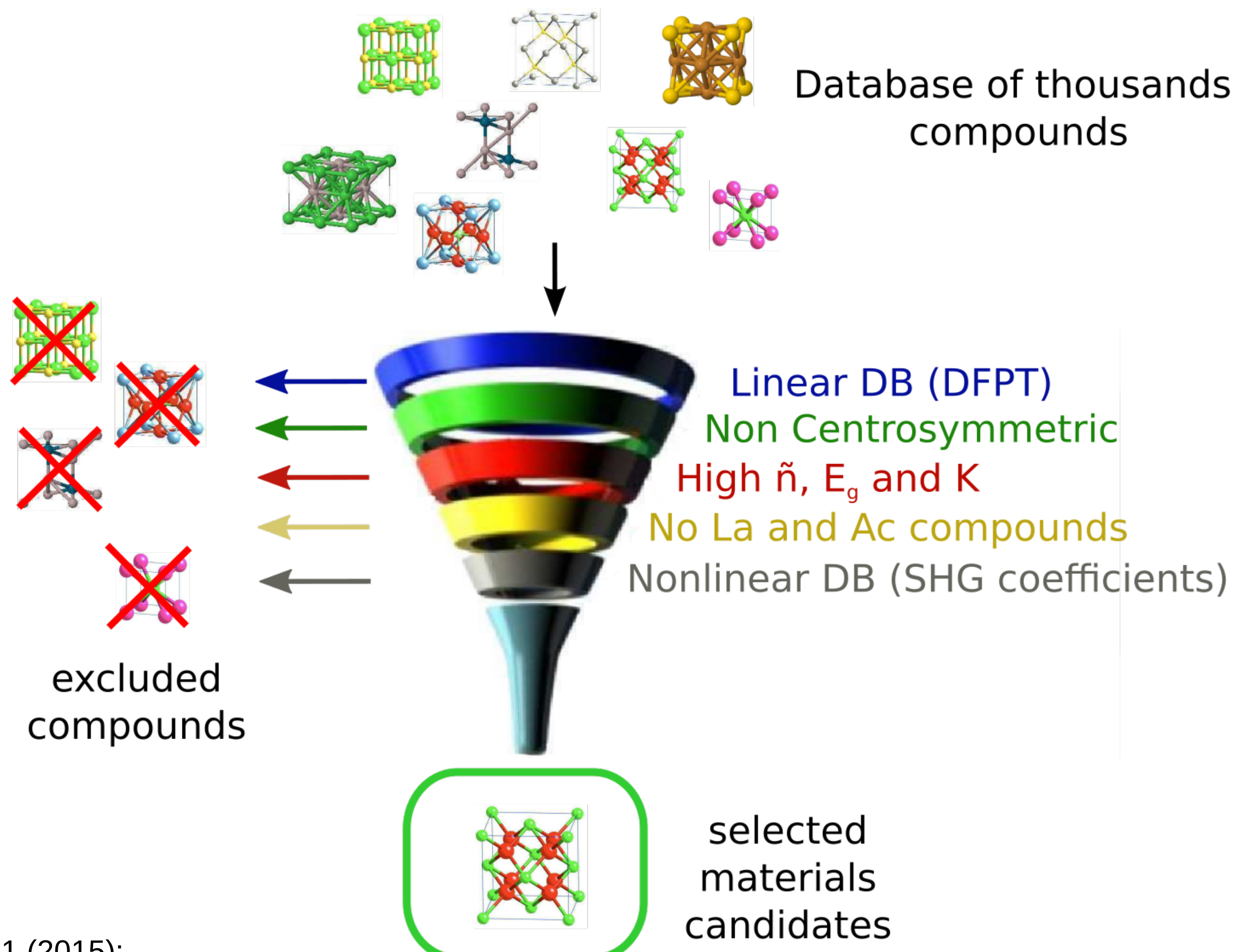


Non-centrosymmetric crystal



High band gap energy





Annu. Rev. Mater. Res., 45, 491 (2015);

THEORETICAL BACKGROUND

$$P_i = P_i^0 + \sum_j \chi_{ij}^{(1)} \varepsilon_j + \sum_{j,l} \chi_{ijl}^{(2)} \varepsilon_j \varepsilon_l + \ldots$$

Linear dielectric susceptibility

$$\chi_{ij}^{(1)} = \frac{1}{\Omega_0} \frac{\partial^2 E}{\partial \varepsilon_i \partial \varepsilon_j}$$

DFPT to compute the 1st order wave function

↓

$$\epsilon_{ij}^{\infty}$$

Macroscopic dielectric tensor

↓

The refractive index is the square root of the macroscopic dielectric tensor

$$\tilde{n} = \sqrt{\epsilon_{ij}^{\infty}}$$

Second order nonlinear susceptibility

$$\chi_{ijl}^{(2)} = \frac{-1}{2\Omega_0} \frac{\partial^3 E}{\partial \varepsilon_i \partial \varepsilon_j \partial \varepsilon_l}$$

2n+1 theorem

If the wave function is known up to the nth order, the energy is known up to (2n+1)th order

↓

$$d_{ijl} = \frac{1}{2} \chi_{ijl}^{(2)}$$

Second order susceptibility tensor

↓

d_{ij} Voigt notation for the second order susceptibility tensor

PRL 58, 1861 (1987); PRB 55, 10335 (1997); PRB 71, 125107 (2005);

METHODOLOGY: WORKFLOW FOR LINEAR AND NONLINEAR OPTICAL PROPERTIES

Linear Database Generation (GGA-PAW)

Relaxed structures from the Materials Project → Calculation of the refractive index with DFPT → Compilation of the DB (ñ, E_g, E_g^d, Space group ...)

↓

Screening of the linear database to extract interesting candidates

Nonlinear Database Generation (LDA-NC)

Nonlinear optical susceptibility for the selected materials →

Authomatized workflow

1. SCF
2. ddk perturbations
3. dde perturbations w.r.t. E. F.
4. dte - only irreducible perturbations w.r.t. E. F.
5. Anaddb → nlflag = 3

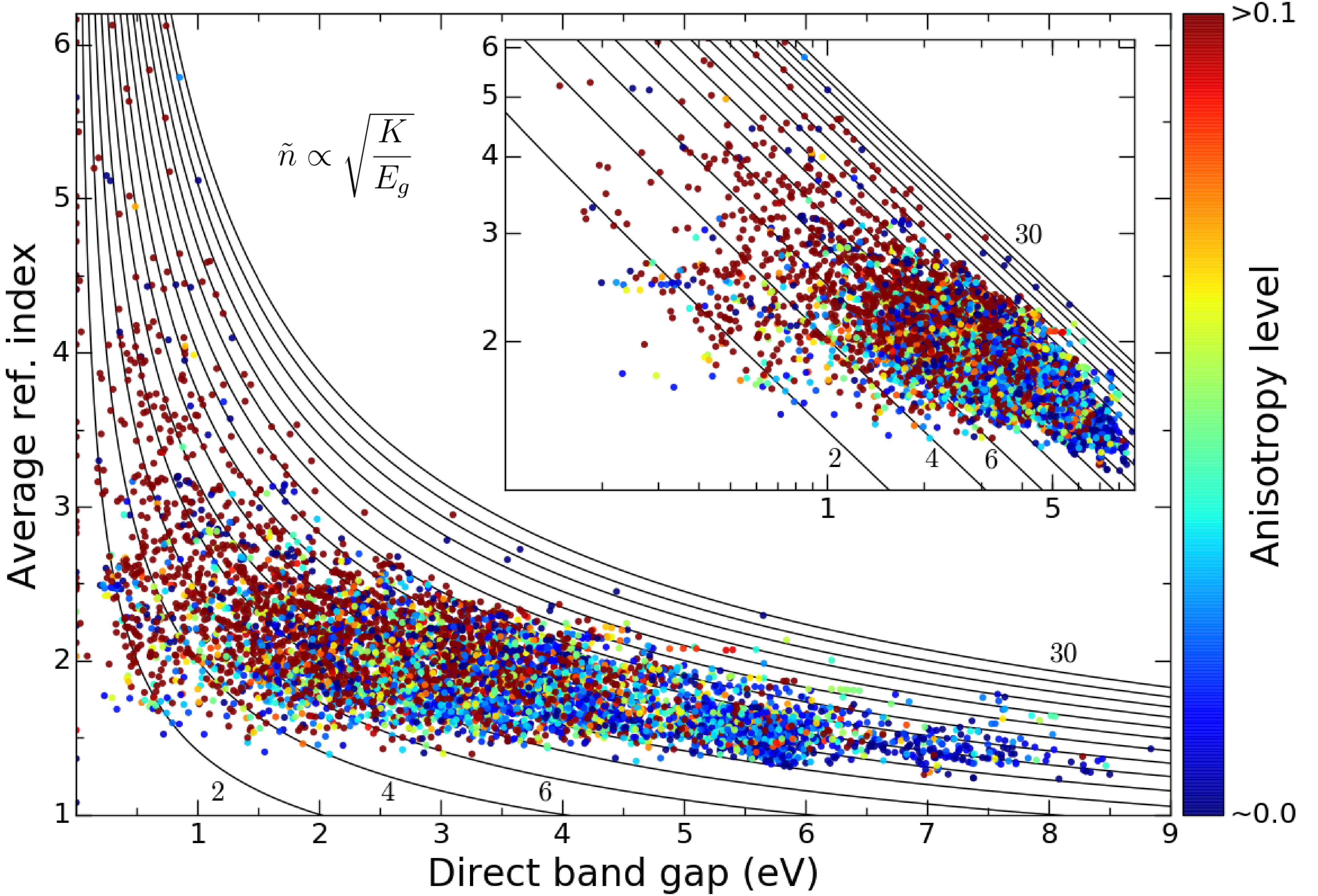
→ (database)

abipy

pymatgen

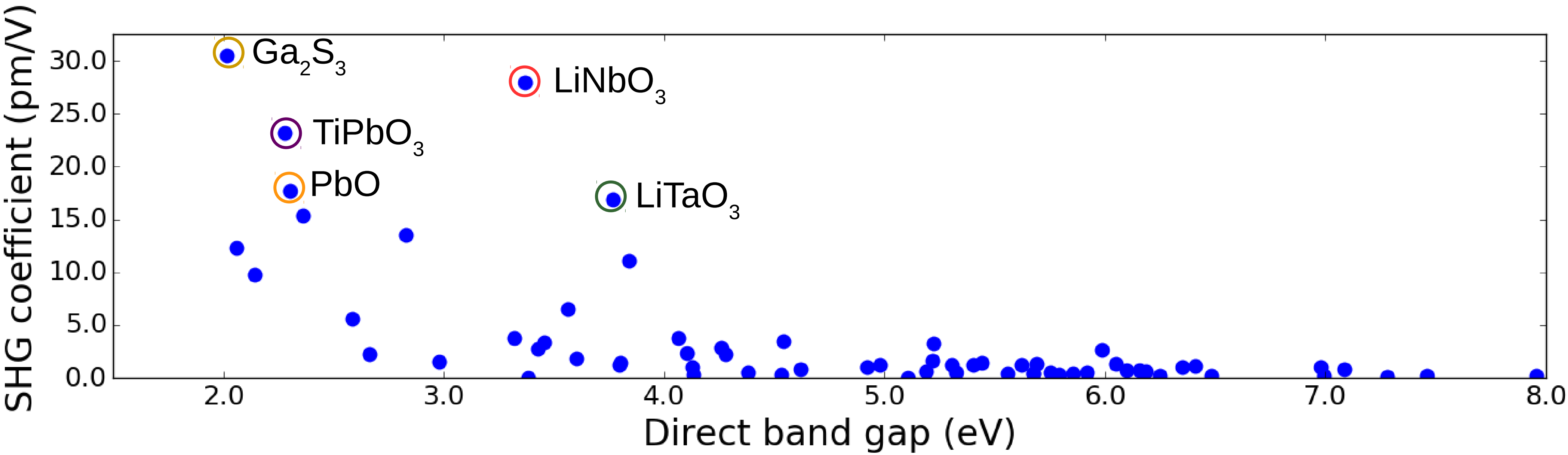
FireWorks

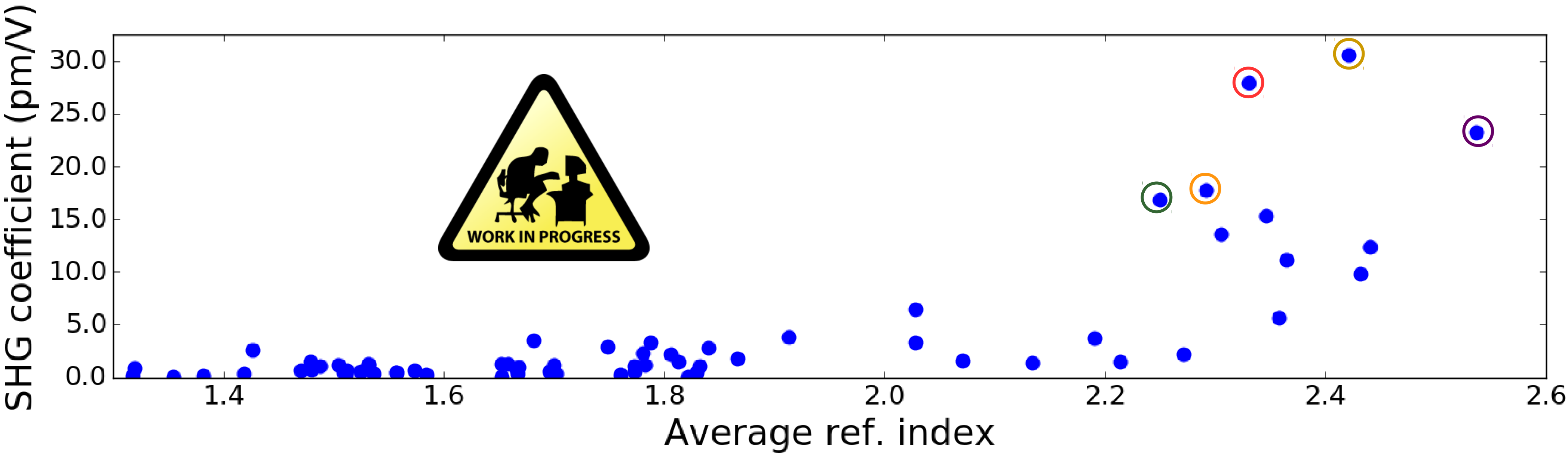
DATABASE IN THE LINEAR REGIME



F. Naccarato, G. Hautier, L. Wirtz, G.-M. Rignanese, in preparation

NONLINEAR DATABASE: PRELIMINARY RESULTS





SUMMARY

PRESENT CONCLUSIONS

- Inverse relation between Band gap and refrective index with some variability in the proportionality constant K
- Inverse relation between SHG coefficient and band gap
- Materials with high linear coefficients show high coefficients in the nonlinear regime

PERSPECTIVES

- Deep analysis of the nonlinear database
- Investigation of interesting candidates for nonlinear optic

THANK YOU FOR YOUR ATTENTION!

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EJD FUNMAT
Marie Skłodowska Curie Actions

UCL
Université catholique de Louvain