Experimental and theoretical investigations of the electronic structure, first and second order optical susceptibilities of some Bismuth-Containing Semiconductors

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1st International Workshop on Bismuth-Containing Semiconductors: Theory, Simulation, and Experiment, University of Michigan

July 14 – 16, 2010

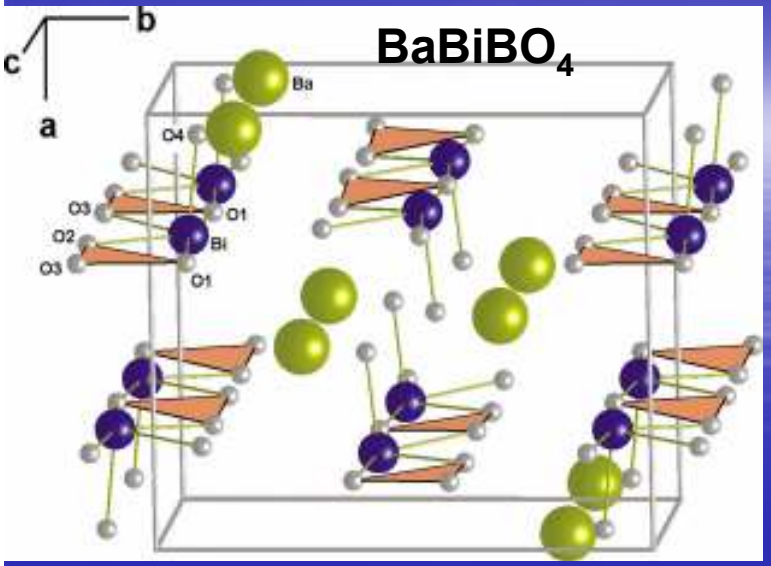
Why is the problem important and why to solve it now

In the recent years, visible, ultraviolet and infrared lasers have been in demand for many industrial, medical, biological and some other important applications.

for this reason ….

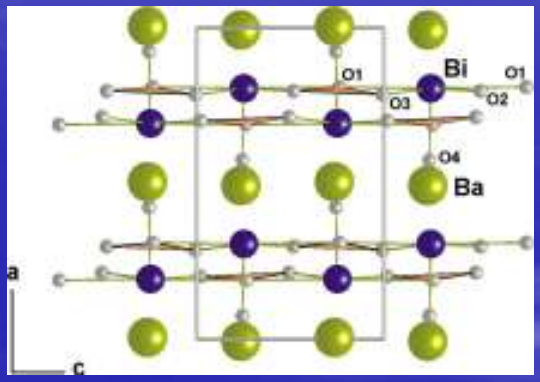
We are doing Theoretical and Experimental research which is concentrated on finding novel NLO materials can achieve our goal.

Crystal structure of Bismuth-Containing Semiconductors



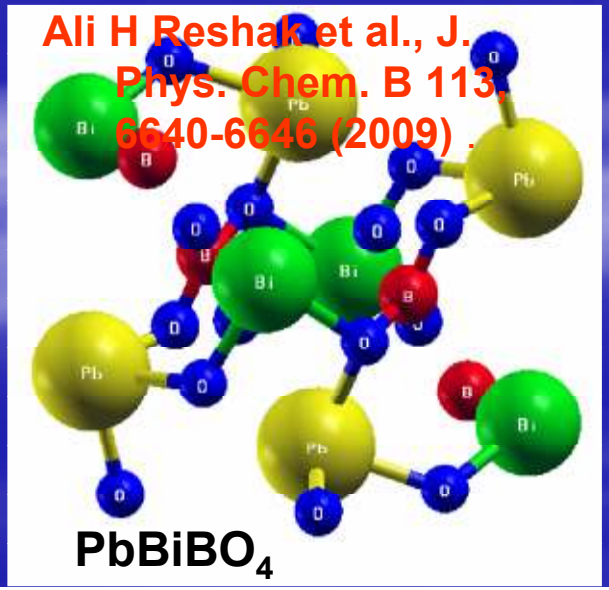
Ali H Reshak et al., J. Solid State Chemistry 181, 789 (2008).

Ali H. Reshak et al., J. Alloys compounds 460, 99-102 (2008)

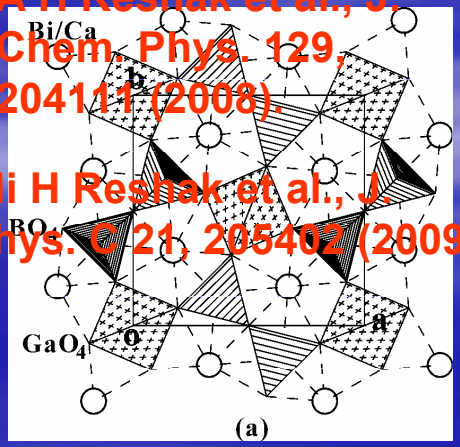


Ali H Reshak et al, Applied Physics A; 91, 451-457 (2008).

Ali H. Reshak et al., J. J. Appl. Phys. 48, 011601 (2009).



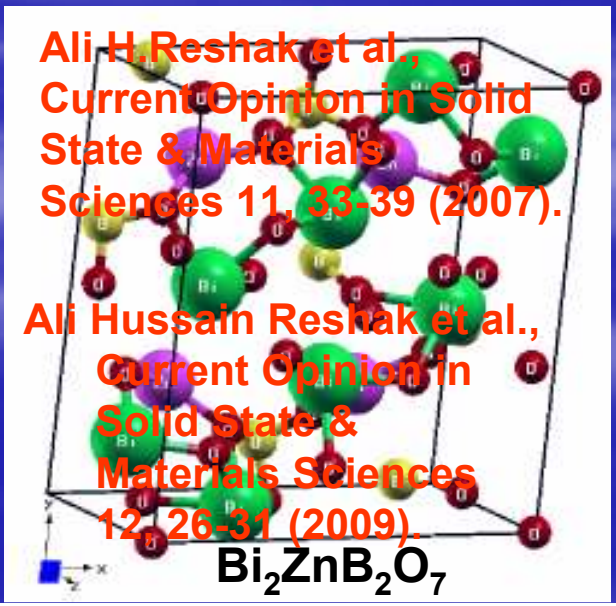
Ali H Reshak et al., J. Phys. Chem. B 113, 6640-6646 (2009) .



CaBiGaB2O7

A H Reshak et al., J. Chem. Phys. 129, 204111 (2008).

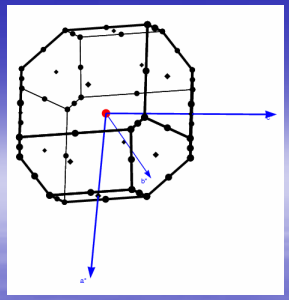
Ali H Reshak et al., J. Phys. C 21, 205402 (2009).



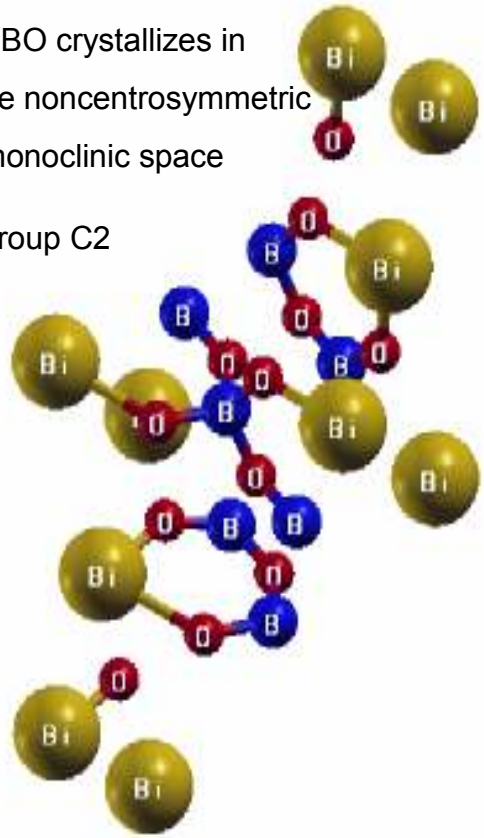
Ali H.Reshak et al., Current Opinion in Solid State & Materials Sciences 11, 33-39 (2007).

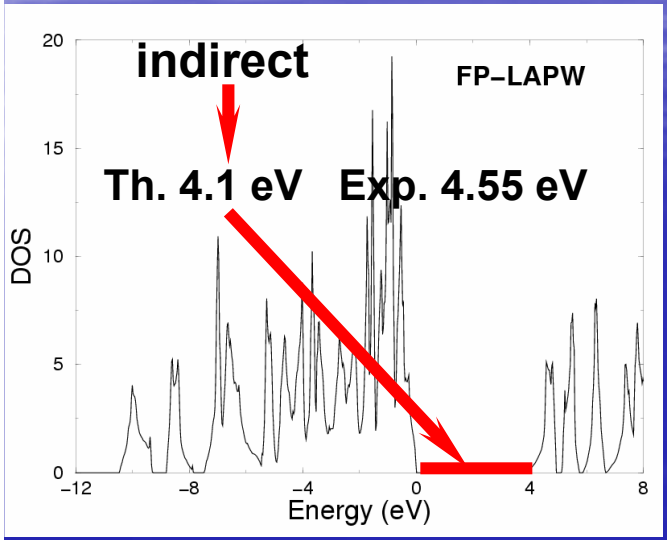
Ali Hussain Reshak et al., Current Opinion in Solid State & Materials Sciences 12, 26-31 (2009).

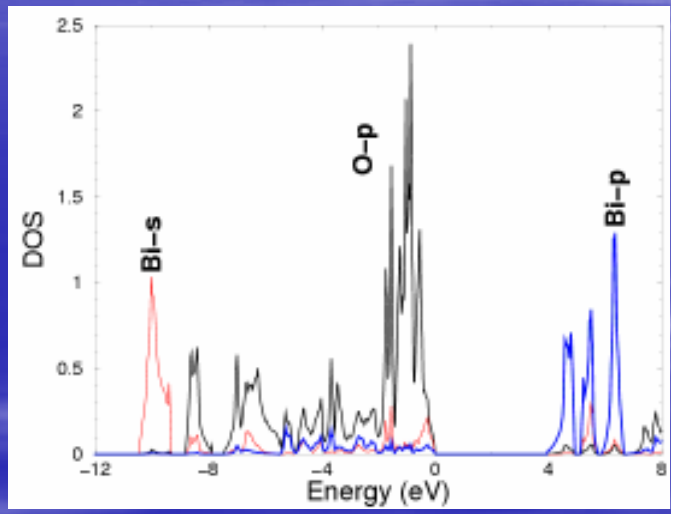
BiB_3O_6

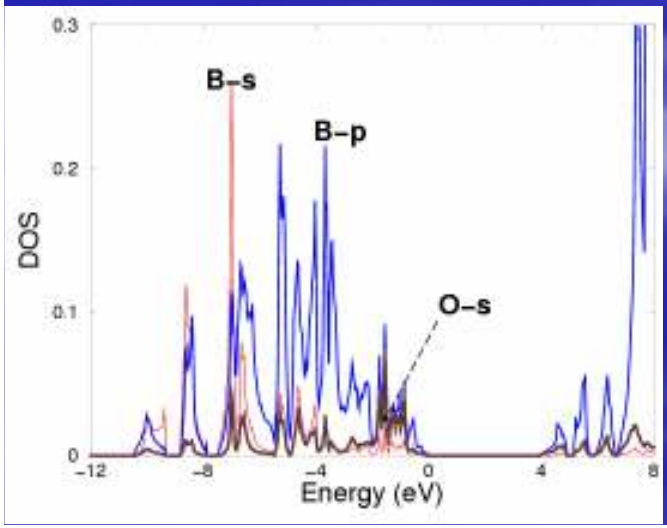


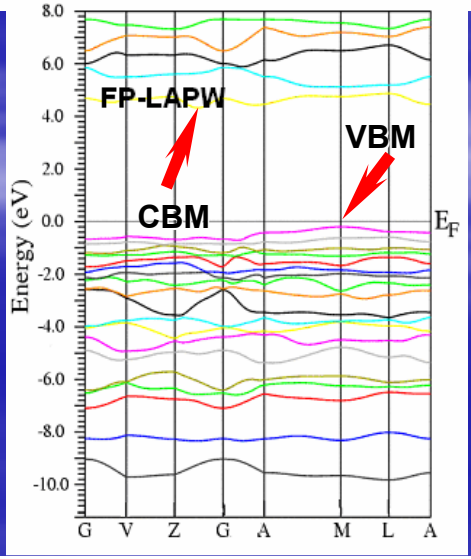
BIBO crystallizes in the noncentrosymmetric monoclinic space group C2

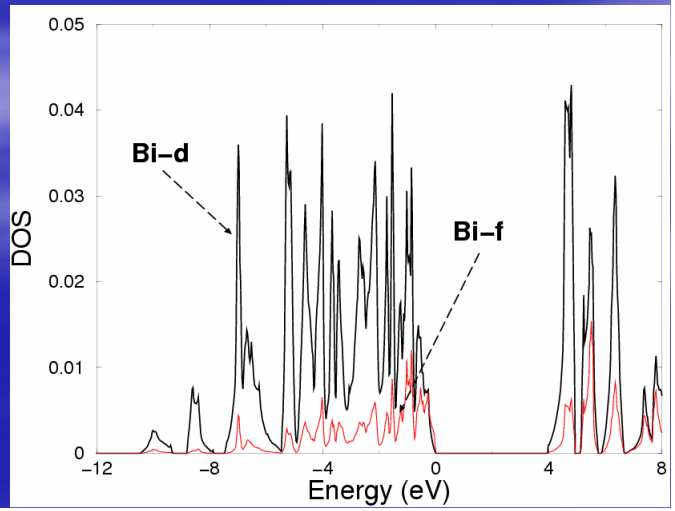




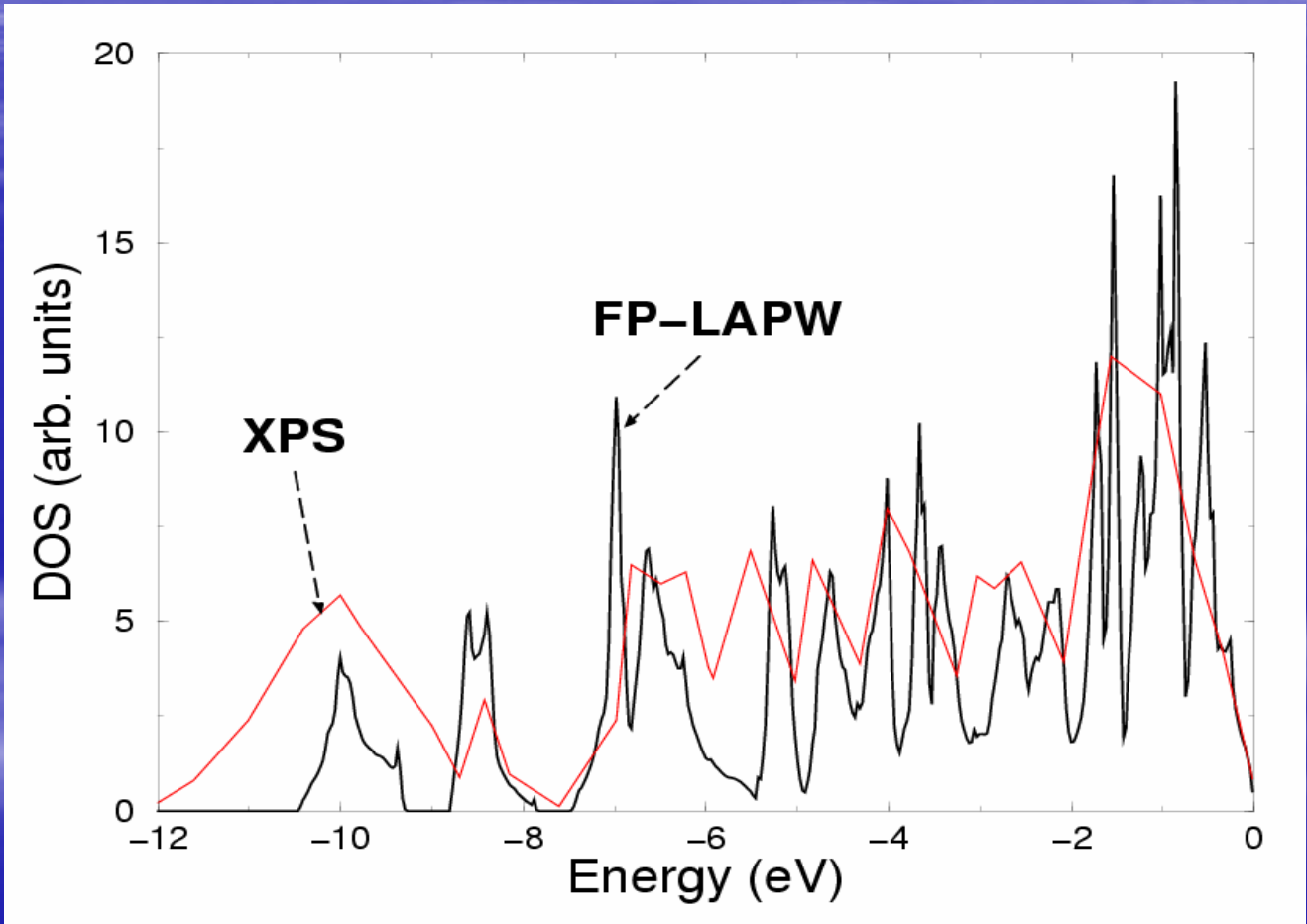


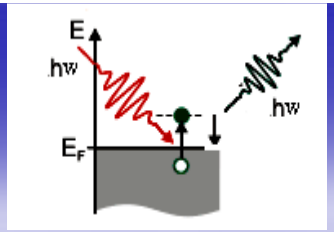




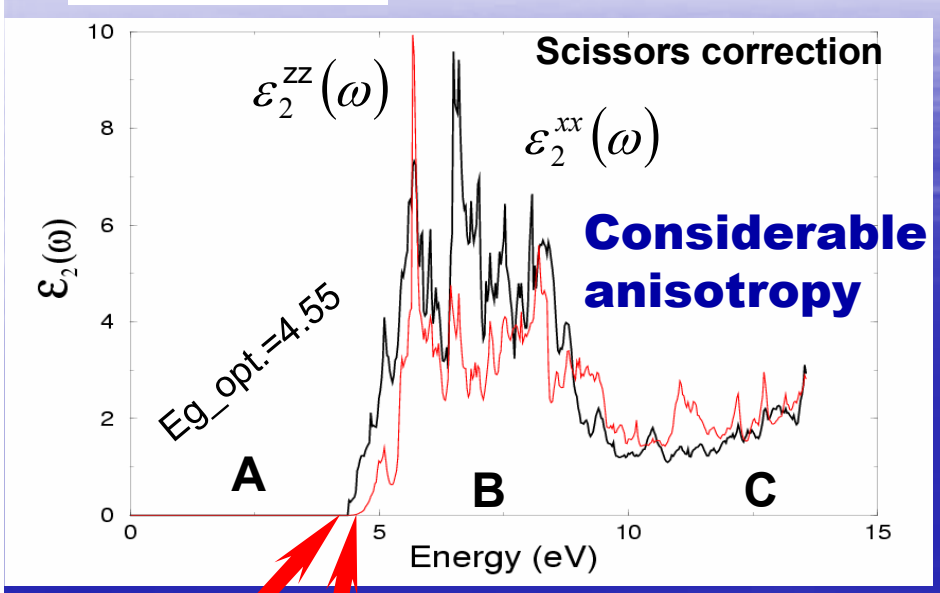


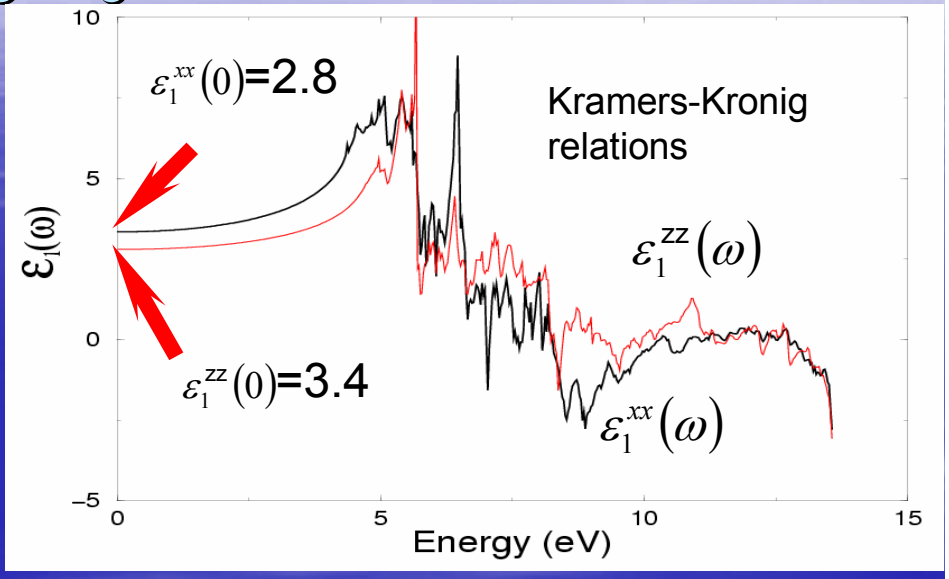
Comparison of the DOS obtained from X-ray Photoelectron Spectra with the electronic structure calculations





Linear optical properties of BiB_3O_6

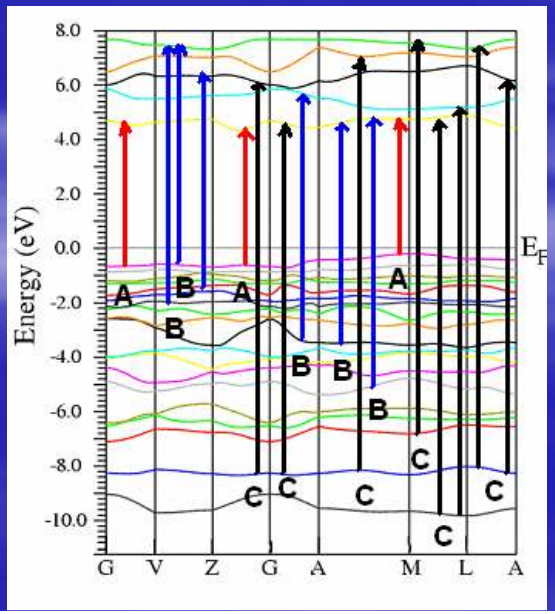




4.4 eV

4.55 eV

Edge of optical Absorption
(First critical point)
(threshold)

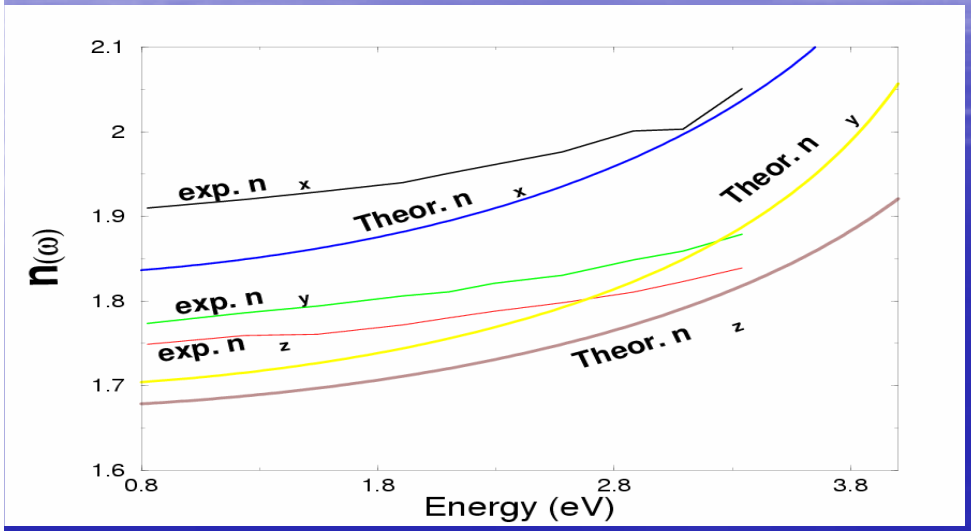


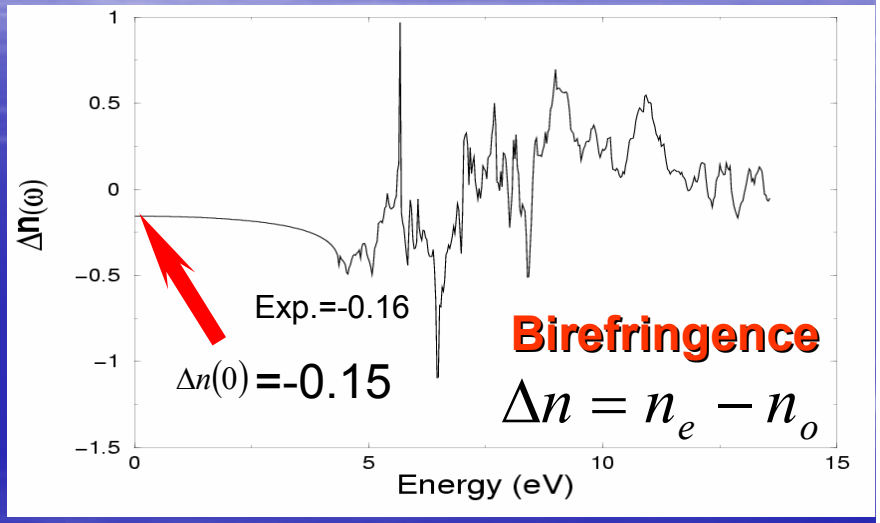
The uniaxial anisotropy

$$\left[\delta\varepsilon = \left(\varepsilon_0^{zz} - \varepsilon_0^{xx}\right)/\varepsilon_0^{tot}\right] = -0.27$$

indicating a strong anisotropy of the dielectric function

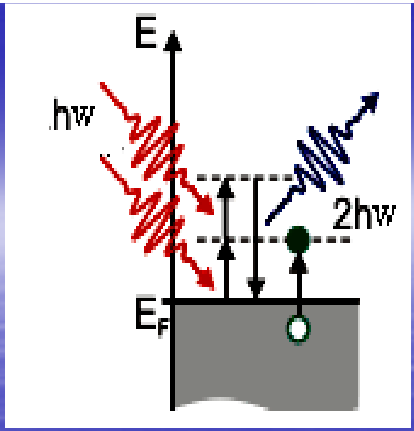
Optical matrix elements





Component	$n(0)$ Theoretical	$n(0)$ experimental
$n_x(0)$	1.83*	1.92*, 1.91[a]
$n_y(0)$	1.70*	1.78*, 1.78[a]
$n_z(0)$	1.67*	1.76*, 1.75[a]

Calculated and measured refractive indices.

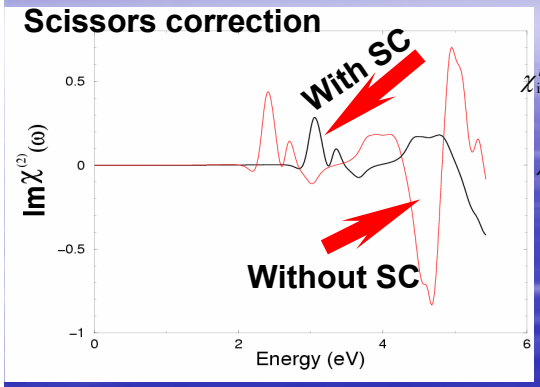


Nonlinear optical properties

- Nonlinear optical techniques are now recognized as the most efficient means available to generate laser radiation at wavelengths that are presently inaccessible via conventional sources.
- In this technology we use nonlinear optical crystals for the frequency conversion of laser light.
- The materials traditionally used for second harmonic generation (SHG) are non-centro-symmetric crystals.

- Since BIBO crystallizes in the non-centrosymmetric monoclinic space group *C*2
- the symmetry allows eight independent nonzero components of the SHG tensor namely; 123, 113, 213, 232, 312, 311, 322 and 333 components (1, 2, and 3 refer to the x, y and z axes, respectively)
- 322 is the dominant component.

Second harmonic generation of BiB_3O_6



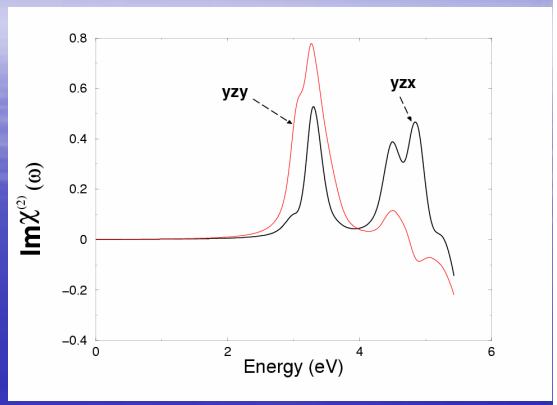
$$\chi^{ijk}_{\text{inter}}(-2\omega;\omega,\omega)=\frac{e^3}{\hbar^2}\sum_{nml}\int\frac{d\vec{k}}{4\pi^3}\frac{\vec{r}^{\,i}_{nm}\left\{\vec{r}^{\,j}_{ml}\vec{r}^{\,k}_{ln}\right\}}{\left(\omega_{ln}-\omega_{ml}\right)}\left\{\frac{2f_{nm}}{\left(\omega_{mn}-2\omega\right)}+\frac{f_{ml}}{\left(\omega_{ml}-\omega\right)}+\frac{f_{ln}}{\left(\omega_{ln}-\omega\right)}\right\}$$

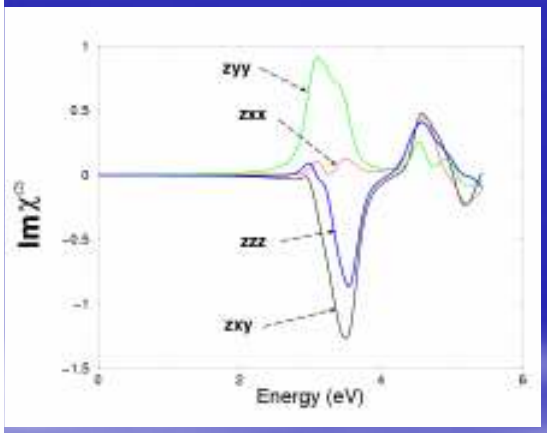
$$\chi^{ijk}_{\text{intra}}(-2\omega;\omega,\omega)=\frac{e^3}{\hbar^2}\int\frac{d\vec{k}}{4\pi^3}\left[\sum_{nml}\omega_{nm}\vec{r}^{\,i}_{nm}\left\{\vec{r}^{\,j}_{ml}\vec{r}^{\,k}_{ln}\right\}\left\{\frac{f_{nl}}{\omega^2_{ln}\left(\omega_{ln}-\omega\right)}-\frac{f_{lm}}{\omega^2_{ml}\left(\omega_{ml}-\omega\right)}\right\}\right.$$

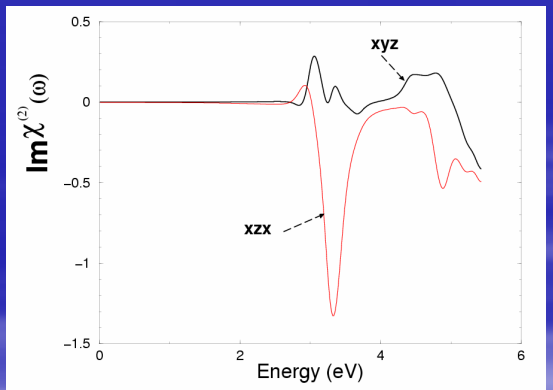
$$\left.-8i\sum_{nm}\frac{f_{nm}\vec{r}^{\,i}_{nm}\left\{\Delta^j_{mn}\vec{r}^{\,k}_{nm}\right\}}{\omega^2_{mn}\left(\omega_{mn}-2\omega\right)}+2\sum_{nml}\frac{f_{nm}\vec{r}^{\,i}_{nm}\left\{\vec{r}^{\,j}_{ml}\vec{r}^{\,k}_{ln}\right\}\left(\omega_{ml}-\omega_{ln}\right)}{\omega^2_{mn}\left(\omega_{mn}-2\omega\right)}\right]$$

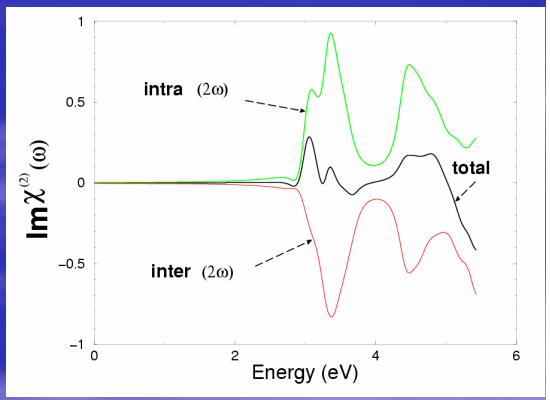
$$\chi^{ijk}_{\text{mod}}(-2\omega;\omega,\omega)=\frac{e^3}{2\hbar^2}\int\frac{d\vec{k}}{4\pi^3}\left[\sum_{nml}\frac{f_{nm}}{\omega^2_{mn}\left(\omega_{mn}-\omega\right)}\left\{\omega_{nl}\vec{r}^{\,i}_{lm}\left\{\vec{r}^{\,j}_{mn}\vec{r}^{\,k}_{nl}\right\}-\omega_{lm}\vec{r}^{\,i}_{nl}\left\{\vec{r}^{\,j}_{lm}\vec{r}^{\,k}_{mn}\right\}\right\}\right.$$

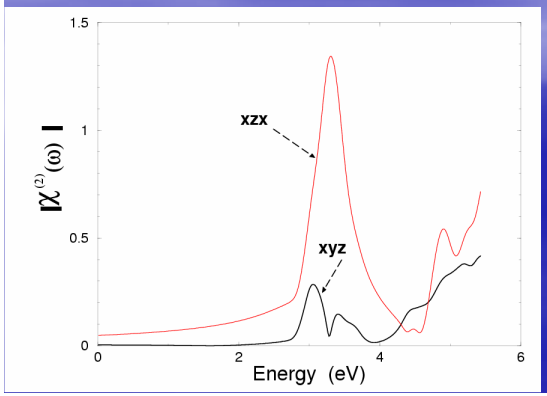
$$\left.-i\sum_{nm}\frac{f_{nm}\vec{r}^{\,i}_{nm}\left\{\vec{r}^{\,j}_{mn}\Delta^k_{mn}\right\}}{\omega^2_{mn}\left(\omega_{mn}-\omega\right)}\right]$$

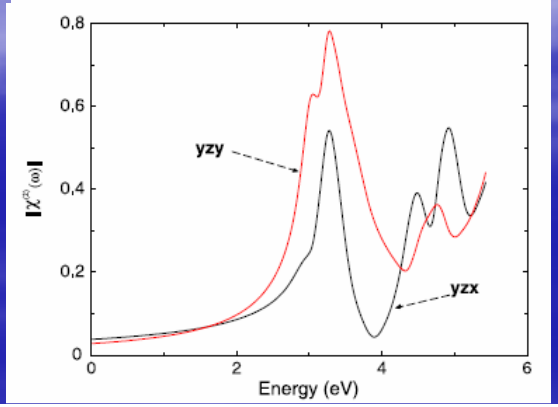


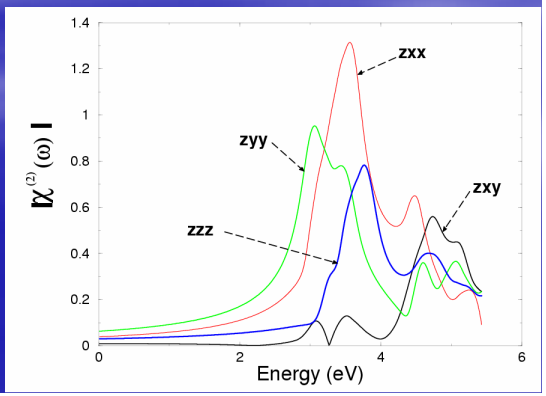












Component	123	131	231	232	311	312	322	333
$Re\,\chi^{abc}(0)total$	-0.005	-0.05	0.04	0.25	0.0	-0.01	0.07	-0.04
$Re\,\chi^{abc}(0)$int *er*	-0.08	-0.105	-0.02	-0.02	-0.4	-0.08	-0.02	-0.1
$Re\,\chi^{abc}(0)$int *ra*	0.1	0.1	0.03	0.05	0.22	0.1	0.12	0.02
Total $Re\,\chi^{abc}(0)$pm/V	-0.2	-2.0	1.8	1.2	-1.8	-0.4	2.8	-1.5

Calculated total and intra- and inter-band of the real part of the $Re\,\chi^{(2)}_{ijk}(\omega)$ at zero frequency in units of $1x10^{-7}$ esu. The *Total* $Re\,\chi^{(2)}_{ijk}(0)$ pm/V is expressed in $1x10^{-12}$ pm/V, in SI units.

Ali Hussain Reshak et al. , Applied Physics A; Materials Science & Processing 91, 451-457 (2008).

$BaBiBO_4$

Ali Hussain Reshak et al., J. Solid State Chemistry 181, 789 (2008).

Calculated total and intra inter-band of the zero frequency of the real part of the $\mathrm{Re}\chi^{(2)}_{ijk}(\omega)$

Components	*xxz*	*yyz*	*zxx*	*zyy*	*zzz*
$\mathrm{Re}\chi_{ijk}(0)$ total	−0.01	0.02	−0.02	0.025	0.01
$\mathrm{Re}\chi_{ijk}(0)$ inter	−0.13	−0.22	−0.22	−0.4	−0.36
$\mathrm{Re}\chi_{ijk}(0)$ intra	0.12	0.2	0.11	0.3	0.25
Total $\mathrm{Re}\chi_{ijk}(0)$ pm/V	0.4	0.55	−0.45	0.54	0.15

The $\mathrm{Re}\chi^{(2)}_{ijk}(0)$ total, inter, and intra are expressed in units of 1×10^{-7} esu. The total $\mathrm{Re}\chi^{(2)}_{ijk}(0)$ pm/V is expressed in pm/V, in SI units.

Components	E_{max} (eV)	I_{max} (in 10^{-7} esu)	χ_{exp} (pm/V)	χ_{calc} (pm/V)
xxz	3.4	0.14	0.51	0.4
yyz	2.7	0.18	1.13	1.08
zzz	3.2	0.13	0.13	0.15

Principal experimental and theoretically calculated components: position of theoretically calculated first spectral maxima (E_{max}); calculated absolute values of the first spectral maxima (in 10^{-7} esu)—I_{max}; experimentally measured values of the second-order susceptibilities at 1064 nm in pm/V—χ_{exp}

Summary

- These crystals possess high Birefringence
- The SHG efficiency of these crystals is about five to ten times larger than KDP (KH_2PO_4).
- The crystals are very stable and reliable for high power and ultrafast lasers

- **Here are some of my publications which are related to the topic of the conference;**
  - Experimental and theoretical investigation of the First and Second order optical susceptibilities of BiB3O6 single crystals, Ali Hussain Reshak , S. Auluck, and I. V. Kityk, Applied Physics A; Materials Science & Processing 91, 451-457 (2008).
  - Linear and Nonlinear optical properties of a novel non centro-symmetric borate oxide BaBiBO4, Ali Hussain Reshak , S. Auluck, and I. V. Kityk, J. Solid State Chemistry 181, 789 (2008).
  - Specific features of second order optical susceptibilities for a complex borate crystal, Bi2ZnB2O7: Experiment and theory, Ali H.Reshak, Xuean Chen, I.V. Kityk and S. Auluck, Current Opinion in Solid State & Materials Sciences 11, 33-39 (2007).
  - Energy band structure and density of states for BaBiBO4 non-linear optical crystal, Ali H. Reshak, I.V. Kityk, S. Auluck, J. Alloys compounds 460, 99-102 (2008)
  - X-ray diffraction and optical properties of a non-centrosymmetric borate CaBiGaB2O7 , A H Reshak, Xuean Chen, S. Auluck, I.V. Kityk , J. Chem. Phys. 129, 204111 (2008).
  - Synthesis, IR, UV-VIS spectra, X-ray diffraction and band structure of a non-centrosymmetric borate CaBiGaB2O7 , Ali Hussain Reshak, Xuean Chen, Fangping Song, I.V. Kityk, and S. Auluck, J. Phys. C 21, 205402 (2009).
  - Comparison of the density of states obtained from the X-ray photoelectron spectra with the electronic structure calculations for α -BiB3O6 . Ali H. Reshak, S. Auluck, A.Majchrowski, I.V. Kityk, J. J. Appl. Phys. 48, 011601 (2009).
  - X-ray diffraction, crystal structure and spectral features of the optical susceptibilities of single crystals of the ternary borate oxide lead bismuth tetraoxide, PbBiBO4, Ali Hussain Reshak, I.V. Kityk, and S. Auluck, Xuean Chen, J. Phys. Chem. B 113, 6640-6646 (2009) .
  - X-ray photoelectron spectrum measurements and theoretical calculations of the electronic band structure for non-centrosymmetric Bi2ZnB2O7 single crystal. Ali Hussain Reshak, Xuean Chen, I.V. Kityk, S. Auluck, K. Iliopoulos, S. Couris, and R. Khenata, Current Opinion in Solid State & Materials Sciences 12, 26-31 (2009).
  - Influence of the Tm+3 concentration on the nonlinear optical effects of the BiB3O6:Tm+3 glass nanoparticle-doped polymer, A Majchrowski, Jean Ebothe, K Ozga, Ivan V Kityk, Ali Hussain Reshak, T Lukasiewicz and Mikhail G Brik, Journal of Physics D: Applied Physics 43, 015103 (2010)

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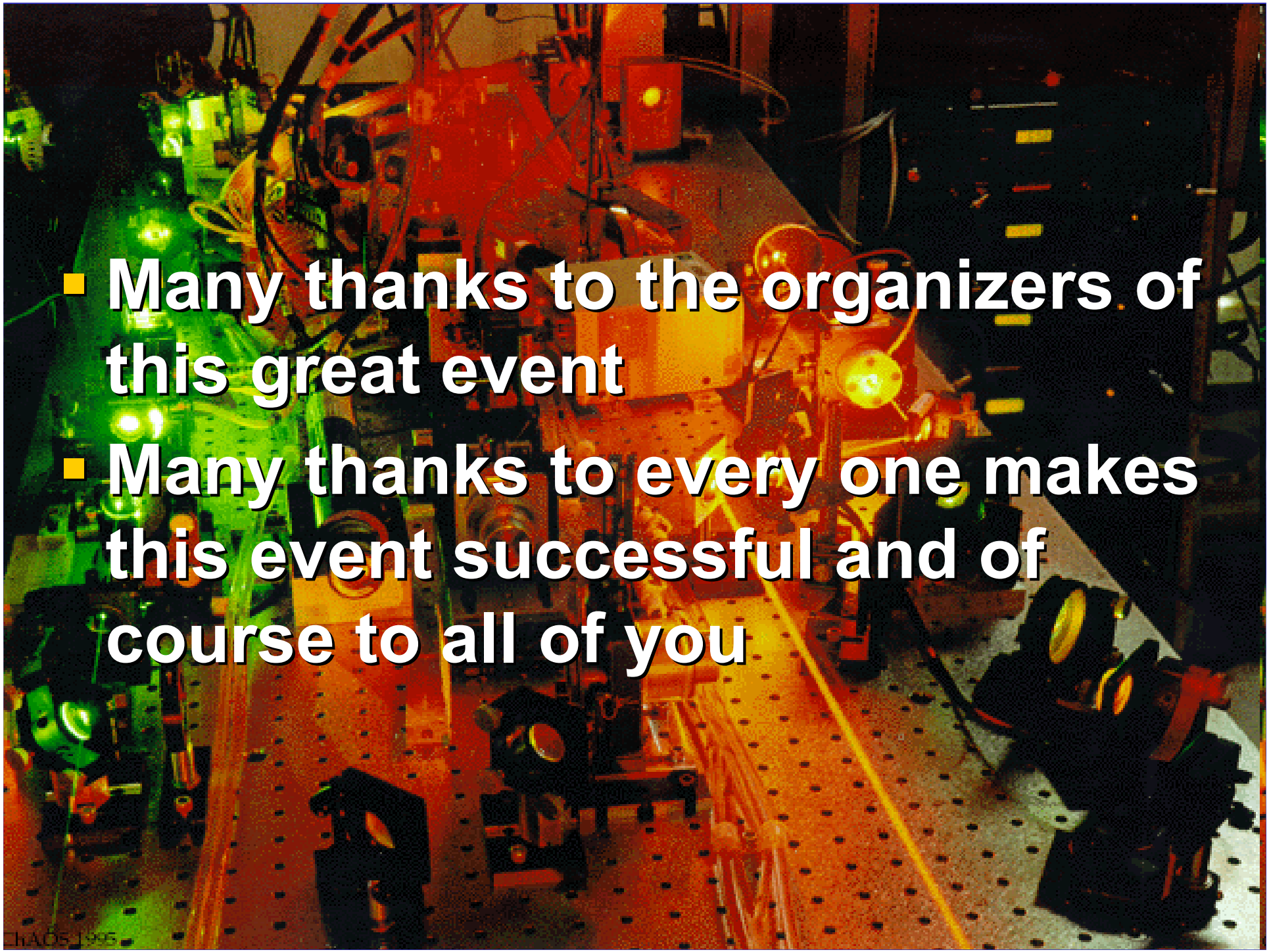

- Many thanks to the organizers of this great event
- Many thanks to every one makes this event successful and of course to all of you