Nonlinear surface magneto-optics of Fe/Cu(001) from first principles: Influences of the Cu substrate

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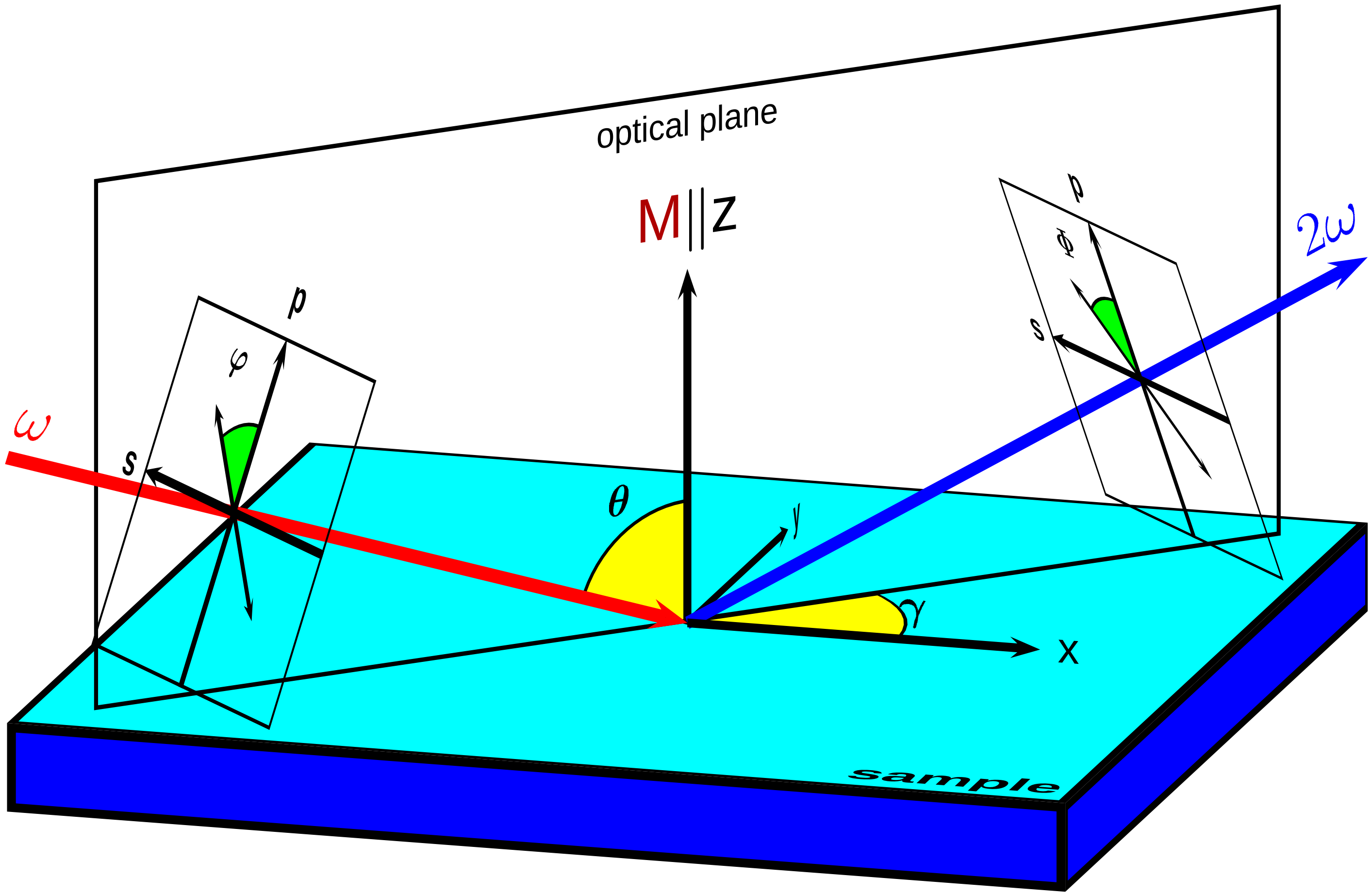
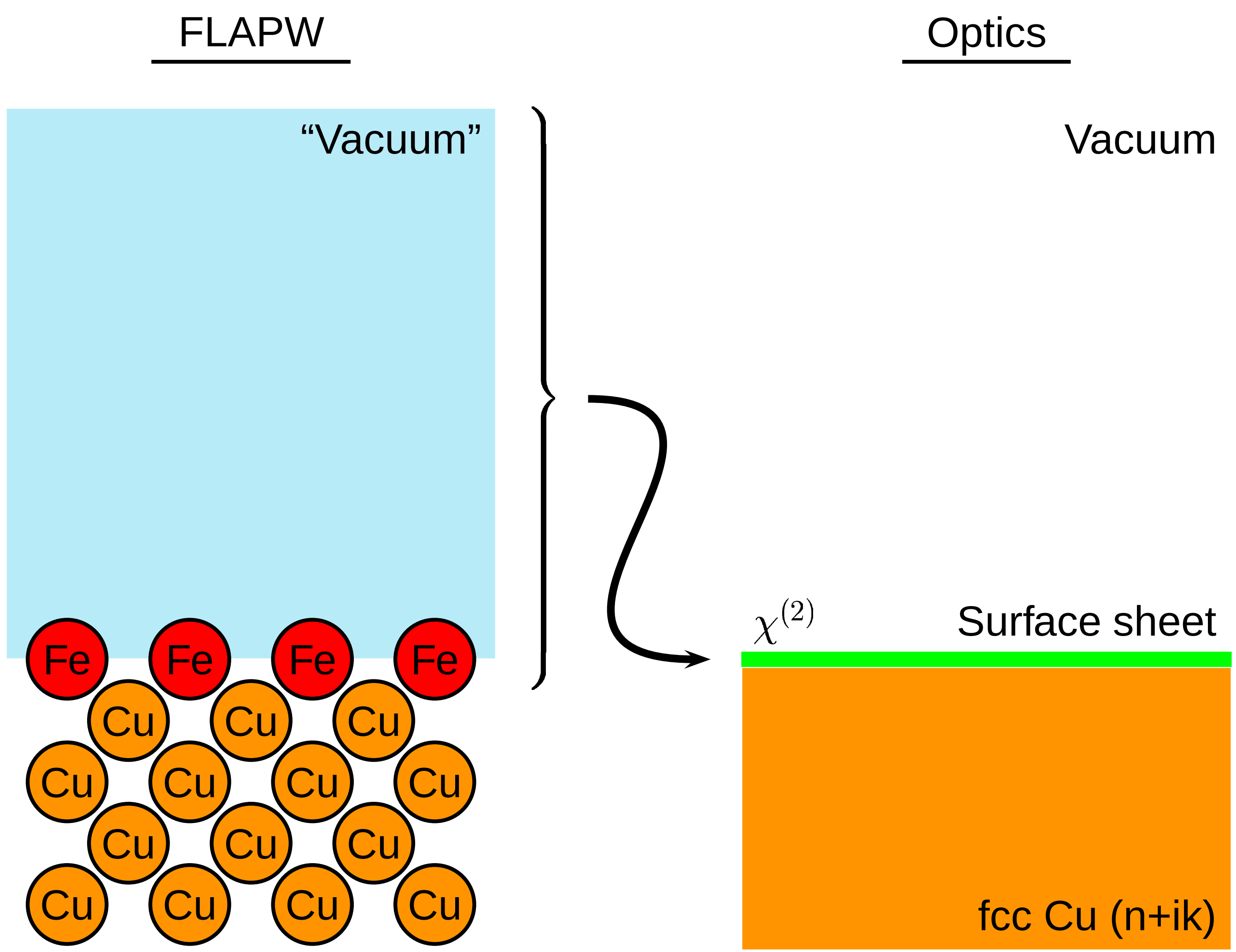
Outline

- Reflection geometry for nonlinear optics
- Numerical calculations: ▶ method ▶ formulas
- Results: ▶ band structure ▶ layer-resolved magnetic moments ▶ screened nonlinear susceptibility ▶ nonlinear optical intensities ▶ rotating the polarizer ▶ nonlinear Kerr rotation
- Conclusions and outlook

Reflection geometry for SHG on magnetic surface



Nonlinear magneto-optics – method



Nonlinear optical field

$$E\left(2\omega;\theta,\Phi,\varphi,\gamma\right)=2i\delta z\frac{\omega}{c}\left|E_0(\omega)\right|^2$$

$$\chi^{(2)}_{ijk}\left(2\mathbf{q},2\omega\right),\ \mathbf{M}\|\mathbf{z}$$

$$\times\left\{A_s\sin\Phi\begin{pmatrix}-\sin\gamma\\ \cos\gamma\\ 0\end{pmatrix}+A_p\cos\Phi\begin{pmatrix}F_c\cos\gamma\\ F_c\sin\gamma\\ N^2F_s\end{pmatrix}\right\}\cdot\left(\begin{array}{ccc|ccc}0&0&0&xyz^-&xxz^+&0\\ 0&0&0&xxz^+&-xyz^-&0\\ zxx^+&zxx^+&zzz^+&0&0&0\end{array}\right)$$

$$\cdot\begin{pmatrix}[t_pf_c\cos\varphi\cos\gamma-t_s\sin\varphi\sin\gamma]^2\\ [t_pf_c\cos\varphi\sin\gamma+t_s\sin\varphi\cos\gamma]^2\\ t_p^2f_s^2\cos^2\varphi\\ 2[t_pf_c\cos\varphi\sin\gamma+t_s\sin\varphi\cos\gamma]t_pf_s\cos\varphi\\ 2[t_pf_c\cos\varphi\cos\gamma-t_s\sin\varphi\sin\gamma]t_pf_s\cos\varphi\\ 2[t_pf_c\cos\varphi\cos\gamma-t_s\sin\varphi\sin\gamma][t_pf_c\cos\varphi\sin\gamma+t_s\sin\varphi\cos\gamma]\end{pmatrix}$$

Screened nonlinear optical susceptibility

$$\chi^{(2)}_{ijk}\left(2\mathbf{q},2\omega\right)=e^3\sum_{\mathbf{k},l,l',l''}\left\{\frac{\langle\mathbf{k}+2\mathbf{q},l''|\mathbf{r}_i|\mathbf{k},l\rangle\langle\mathbf{k},l|\mathbf{r}_j|\mathbf{k}+\mathbf{q},l'\rangle\langle\mathbf{k}+\mathbf{q},l'|\mathbf{r}_k|\mathbf{k}+2\mathbf{q},l''\rangle}{E_{\mathbf{k}+2\mathbf{q},l''}-E_{\mathbf{k},l}-2\hbar\omega+2i\hbar\alpha}\right.$$

$$\left.\times\left(\frac{f(E_{\mathbf{k}+2\mathbf{q},l''})-f(E_{\mathbf{k}+\mathbf{q},l'})}{E_{\mathbf{k}+2\mathbf{q},l''}-E_{\mathbf{k}+\mathbf{q},l'}-\hbar\omega+i\hbar\alpha}-\frac{f(E_{\mathbf{k}+\mathbf{q},l'})-f(E_{\mathbf{k},l})}{E_{\mathbf{k}+\mathbf{q},l'}-E_{\mathbf{k},l}-\hbar\omega+i\hbar\alpha}\right)\right\}$$

$$\times\left[1+4\pi\mathrm{e}^2\sum_{ab}m_am_b\sum_{\mathbf{k},l,l''}\langle\mathbf{k},l\,|\mathbf{r}_a|\,\mathbf{k}+2\mathbf{q},l''\rangle\langle\mathbf{k}+2\mathbf{q},l''\,|\mathbf{r}_b|\,\mathbf{k},l\rangle\frac{f(E_{\mathbf{k}+2\mathbf{q},l''})-f(E_{\mathbf{k},l})}{E_{\mathbf{k}+2\mathbf{q},l''}-E_{\mathbf{k}l}-2\hbar\omega+2i\hbar\alpha}\right]^{-1}$$

Nonlinear intensity

$$I\left(2\omega;\theta,\Phi,\varphi,\gamma\right)=\epsilon_0c_0\left|E\left(2\omega;\theta,\Phi,\varphi,\gamma\right)\right|^2$$

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Details of the calculation

- WIEN97 spin-polarized scf cycle
  - energy convergence to 0.1 mRy
  - no inversion symmetry (surface)
- add magnetization direction
- our implementation of the spin-orbit coupling
  - second variation
  - spherical potential, inside MT sphere only
  - WIEN97 basis set
  - added after complete scf-cycle
- transition matrix elements $\langle \mathbf{k}, l | \mathbf{r}_i | \mathbf{k}', l' \rangle$
- screened nonlinear susceptibility $\chi^{(2)}_{ijk}$
- optical properties (E, I, $\mathcal{A}$, ϕ_K)

Parameters in WIEN97 scf cycle

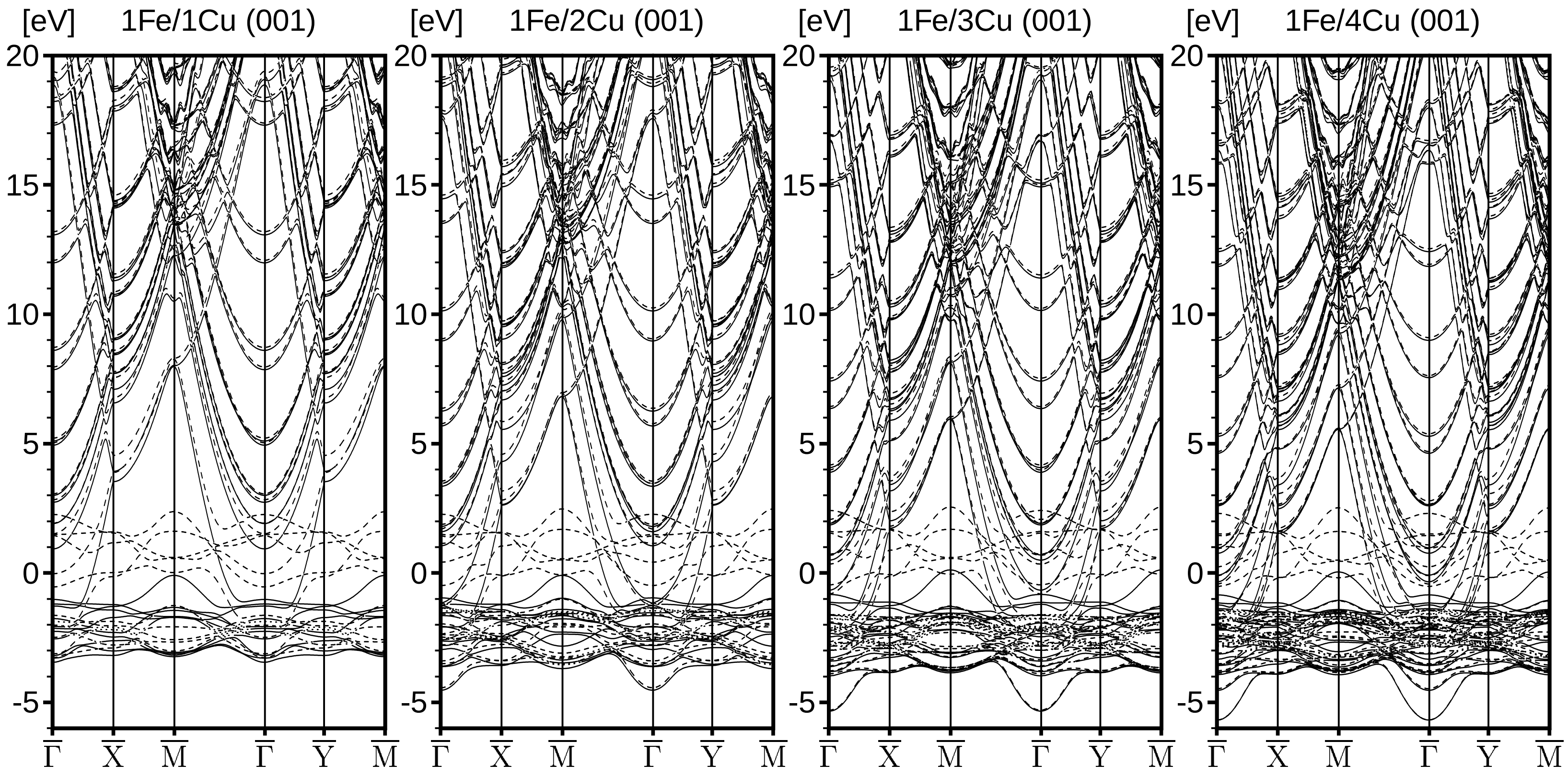
- fcc Cu lattice, $a = 3.61$ Å
- separation 8 vacuum layers
- muffin-tin radius 1.21 Å
- 2502 $\mathbf{k}$-points in I2BZ

Parameters in optics

- $|E_0(\omega)| = 10^8$ V/m
- $\delta z = 4.82$ Å
- $\gamma = 0$, $\theta = \pi/4$
- linear Cu $n + ik$ from [1]

[1] D. W. Lynch and W. R. Hunter, in *Handbook of optical constants in solids*, edited by E. D. Palik (Academic Press, London, 1985), pp. 275–367.

Fe/Cu (001) Band structure with spin-orbit coupling



Solid lines: $>$ 90% majority spin
Dashed lines: $>$ 90% minority spin
Dotted lines: $<$ 90% of either spin

Magnetic spin moments

1 ML Fe on up to 7 ML fcc Cu (001)

Fe	Cu layer number						
	1	2	3	4	5	6	7
2.828	0.046						
2.844	0.043	-0.026					
2.793	0.033	-0.025	-0.003				
2.792	0.048	-0.015	-0.006	-0.065			
2.853	0.042	-0.016	-0.000	0.001	0.001		
2.835	0.044	-0.016	-0.002	-0.001	-0.002	-0.006	
2.740	0.045	-0.016	0.004	0.005	0.008	0.001	-0.010
	FM	AFM					

First Cu layer FM coupled to Fe layer

Second Cu layer AFM coupled to first Cu layer

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Nonlinear optical field

$$E\left(2\omega;\theta,\Phi,\varphi,\gamma\right)=2i\delta z\frac{\omega}{c}\left|E_0(\omega)\right|^2$$

$$\times\left\{A_s\sin\Phi\begin{pmatrix}-\sin\gamma\\ \cos\gamma\\ 0\end{pmatrix}+A_p\cos\Phi\begin{pmatrix}F_c\cos\gamma\\ F_c\sin\gamma\\ N^2F_s\end{pmatrix}\right\}\cdot\left(\begin{array}{ccc|ccc}0&0&0&xyz^-&xxz^+&0\\ 0&0&0&xxz^+&-xyz^-&0\\ zxx^+&zxx^+&zzz^+&0&0&0\end{array}\right)$$

$\chi^{(2)}_{ijk}\left(2\mathbf{q},2\omega\right)$, **M||z**

$$\cdot\begin{pmatrix}[t_pf_c\cos\varphi\cos\gamma-t_s\sin\varphi\sin\gamma]^2\\ [t_pf_c\cos\varphi\sin\gamma+t_s\sin\varphi\cos\gamma]^2\\ t_p^2f_s^2\cos^2\varphi\\ 2[t_pf_c\cos\varphi\sin\gamma+t_s\sin\varphi\cos\gamma]t_pf_s\cos\varphi\\ 2[t_pf_c\cos\varphi\cos\gamma-t_s\sin\varphi\sin\gamma]t_pf_s\cos\varphi\\ 2[t_pf_c\cos\varphi\cos\gamma-t_s\sin\varphi\sin\gamma][t_pf_c\cos\varphi\sin\gamma+t_s\sin\varphi\cos\gamma]\end{pmatrix}$$

Screened nonlinear optical susceptibility

$$\chi^{(2)}_{ijk}\left(2\mathbf{q},2\omega\right)=e^3\sum_{\mathbf{k},l,l',l''}\left\{\frac{\langle\mathbf{k}+2\mathbf{q},l''|\mathbf{r}_i|\mathbf{k},l\rangle\langle\mathbf{k},l|\mathbf{r}_j|\mathbf{k}+\mathbf{q},l'\rangle\langle\mathbf{k}+\mathbf{q},l'|\mathbf{r}_k|\mathbf{k}+2\mathbf{q},l''\rangle}{E_{\mathbf{k}+2\mathbf{q},l''}-E_{\mathbf{k},l}-2\hbar\omega+2i\hbar\alpha}\right.$$

$$\left.\times\left(\frac{f(E_{\mathbf{k}+2\mathbf{q},l''})-f(E_{\mathbf{k}+\mathbf{q},l'})}{E_{\mathbf{k}+2\mathbf{q},l''}-E_{\mathbf{k}+\mathbf{q},l'}-\hbar\omega+i\hbar\alpha}-\frac{f(E_{\mathbf{k}+\mathbf{q},l'})-f(E_{\mathbf{k},l})}{E_{\mathbf{k}+\mathbf{q},l'}-E_{\mathbf{k},l}-\hbar\omega+i\hbar\alpha}\right)\right\}$$

$$\times\left[1+4\pi e^2\sum_{ab}m_am_b\sum_{\mathbf{k},l,l''}\langle\mathbf{k},l\left|\mathbf{r}_a\right|\mathbf{k}+2\mathbf{q},l''\rangle\langle\mathbf{k}+2\mathbf{q},l''\left|\mathbf{r}_b\right|\mathbf{k},l\rangle\frac{f(E_{\mathbf{k}+2\mathbf{q},l''})-f(E_{\mathbf{k},l})}{E_{\mathbf{k}+2\mathbf{q},l''}-E_{\mathbf{k}l}-2\hbar\omega+2i\hbar\alpha}\right]^{-1}$$

Nonlinear intensity

$$I\left(2\omega;\theta,\Phi,\varphi,\gamma\right)=\epsilon_0c_0\left|E\left(2\omega;\theta,\Phi,\varphi,\gamma\right)\right|^2$$

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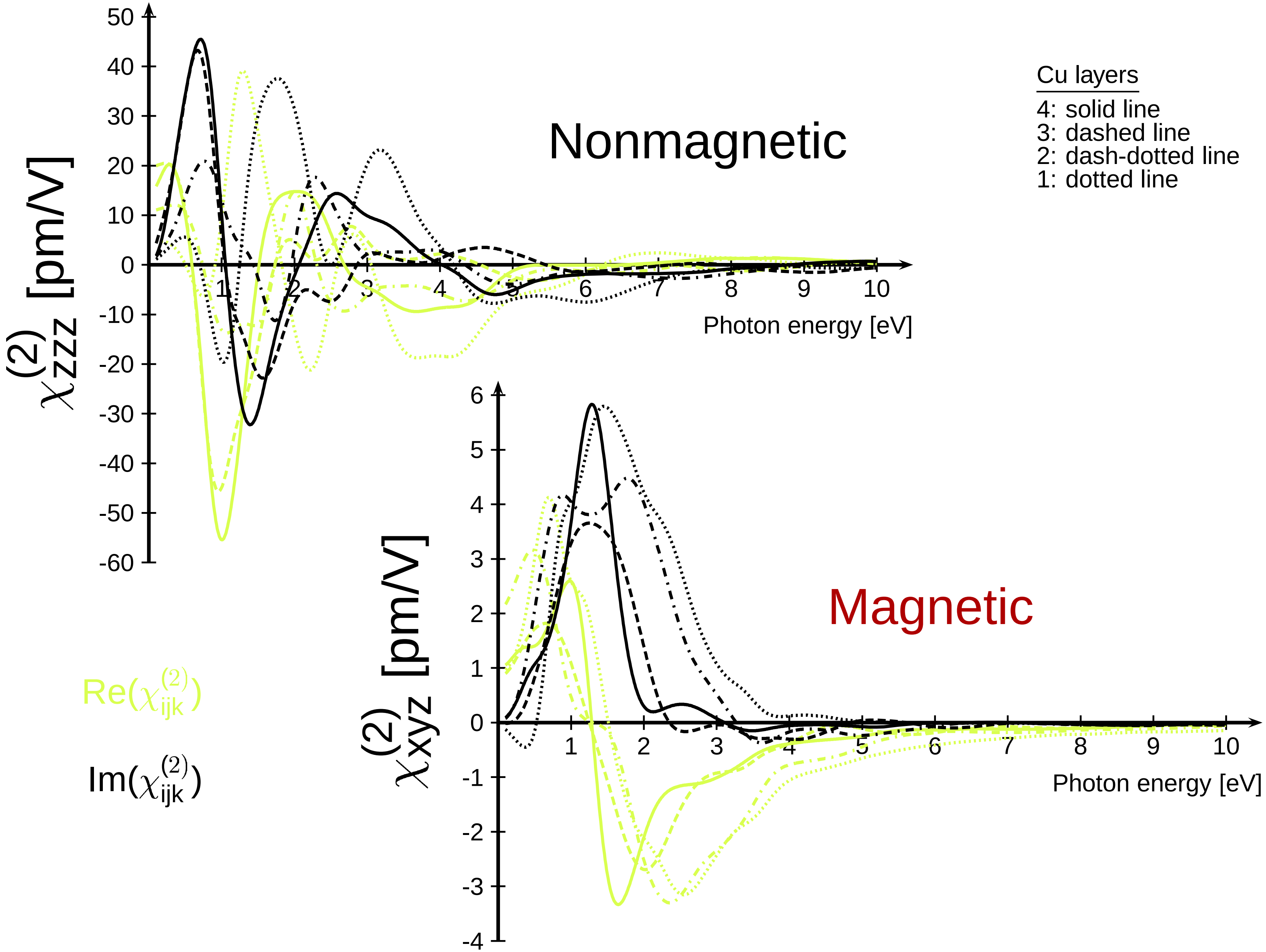
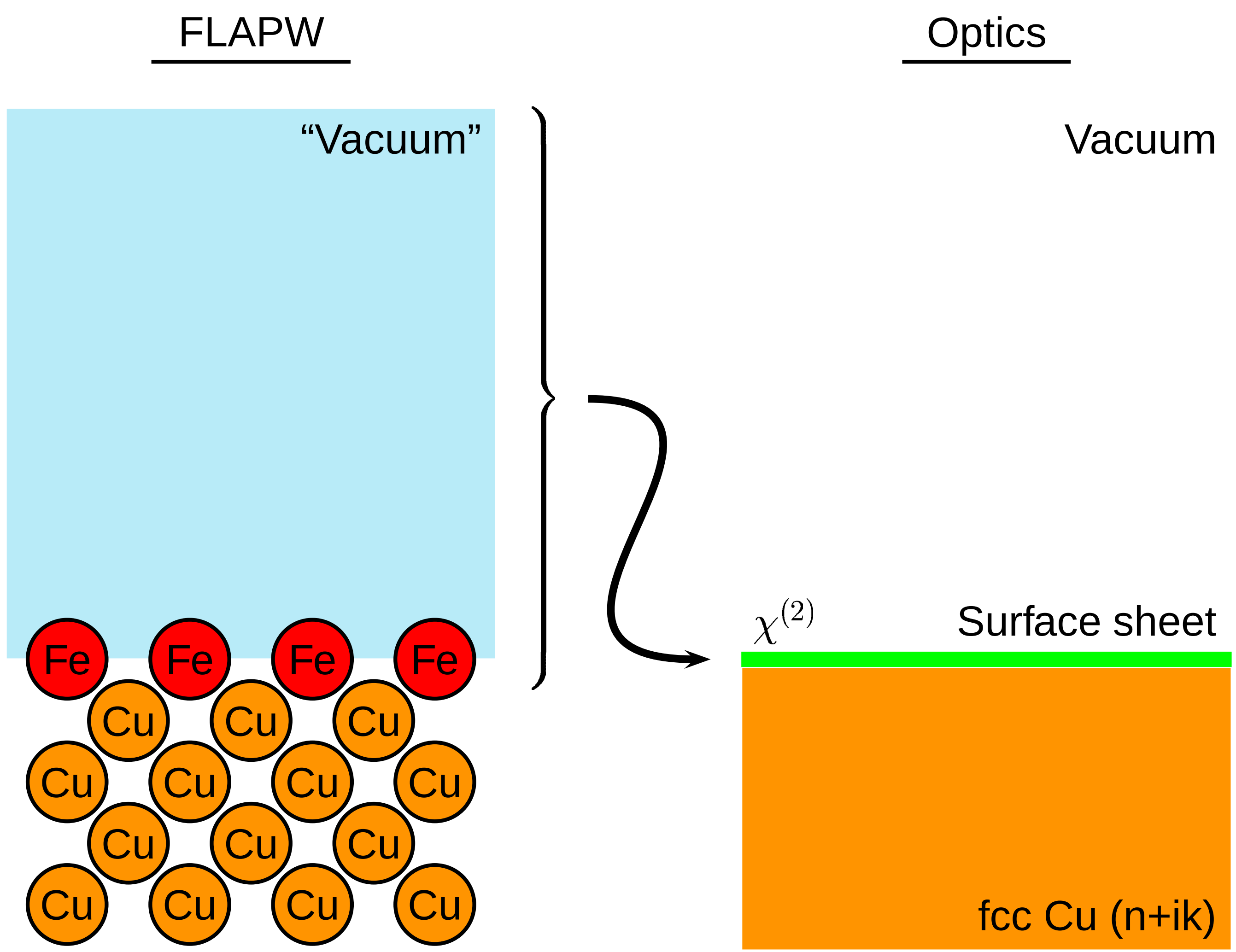
Screened nonlinear susceptibility tensor

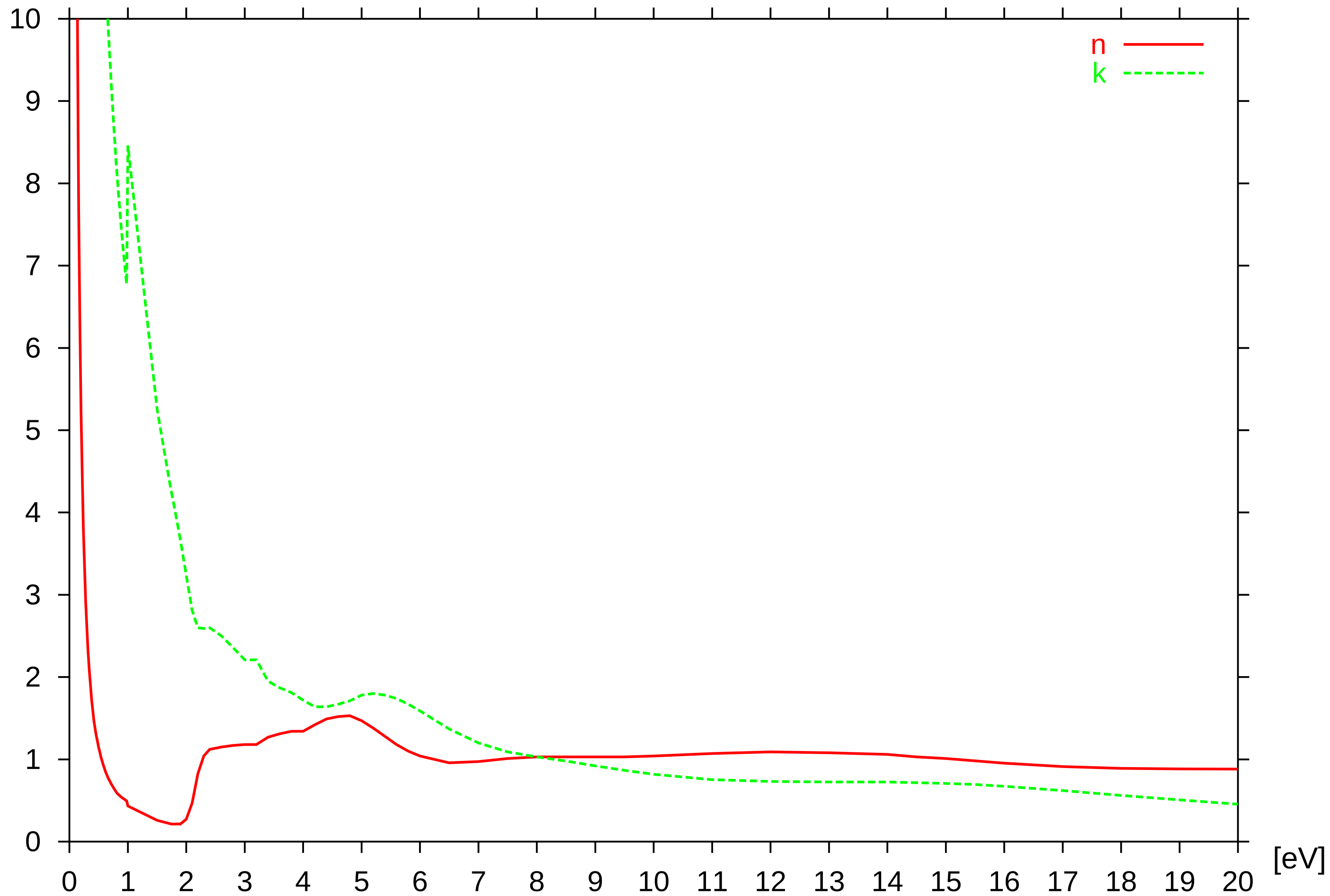


Nonlinear magneto-optics – method



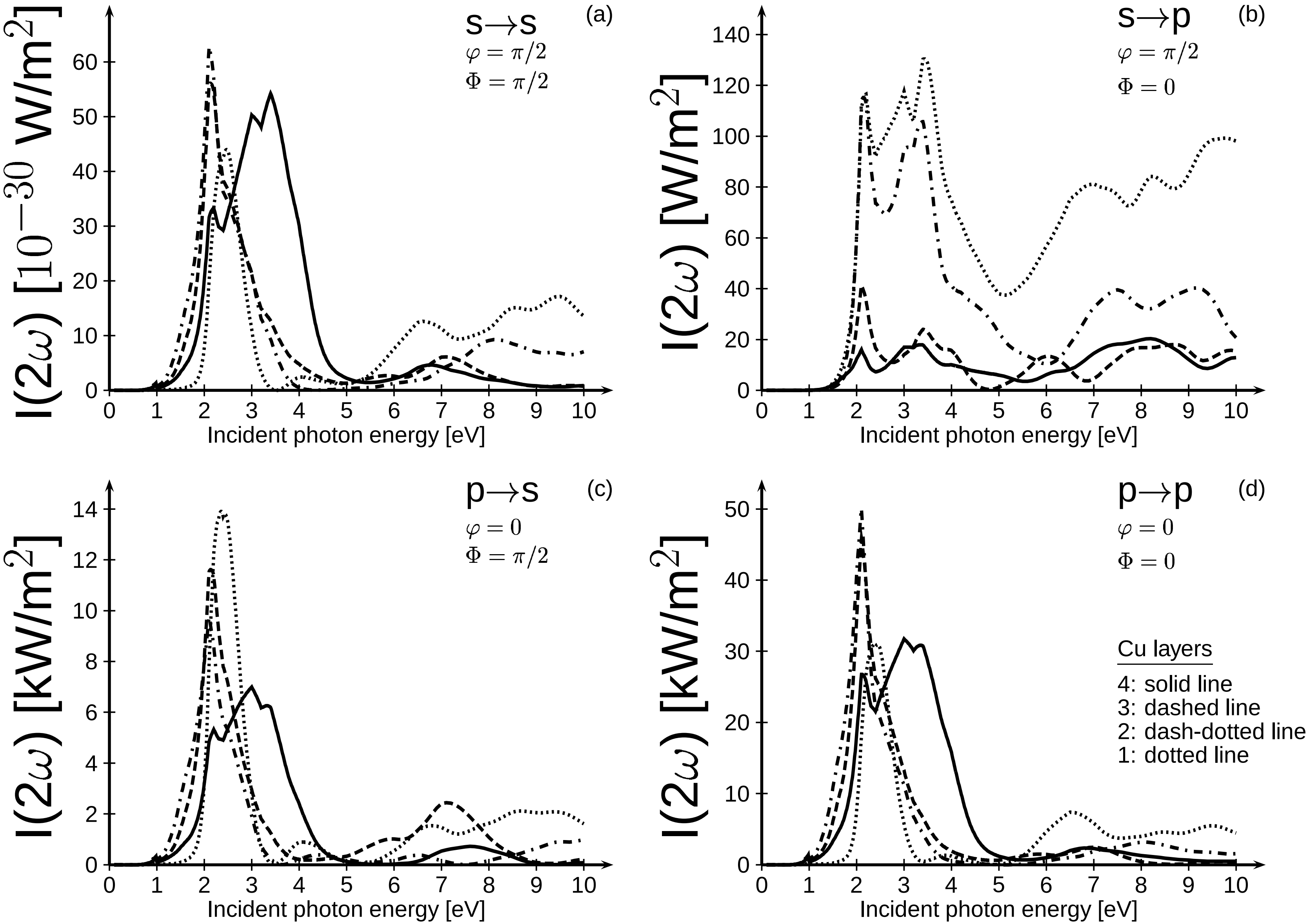
Complex refractive index (n+ik) of Cu

Experimental values



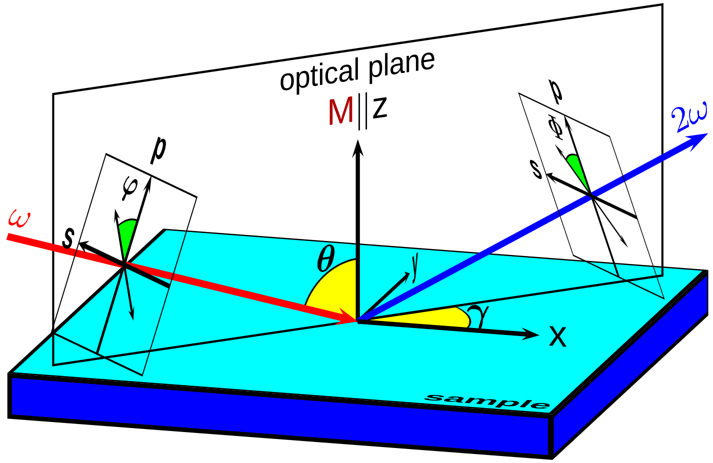
[D. W. Lynch and W. R. Hunter, in *Handbook of optical constants in solids*, edited by E. D. Palik (Academic Press, London, 1985), pp. 275–367]

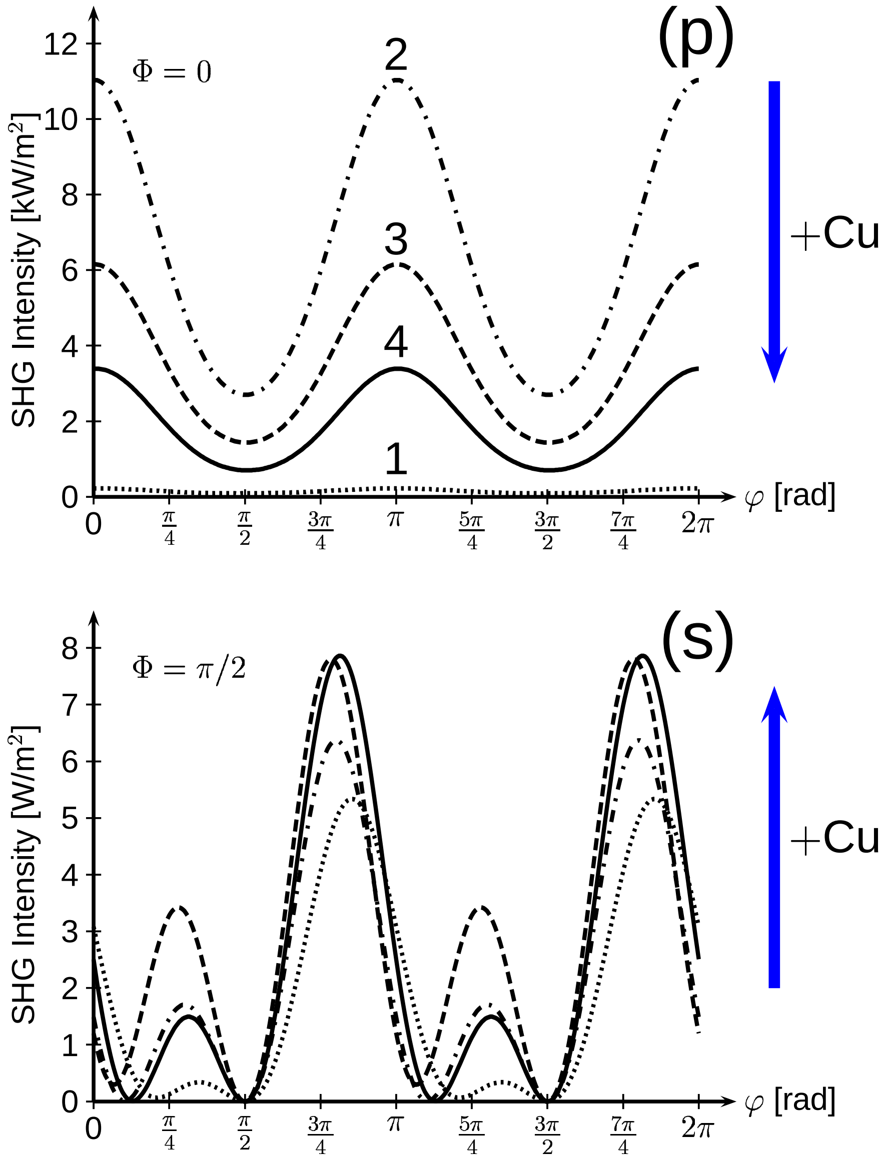
Second-harmonic intensities



Varying the polarizer angle φ

$\hbar\omega$=1.5 eV





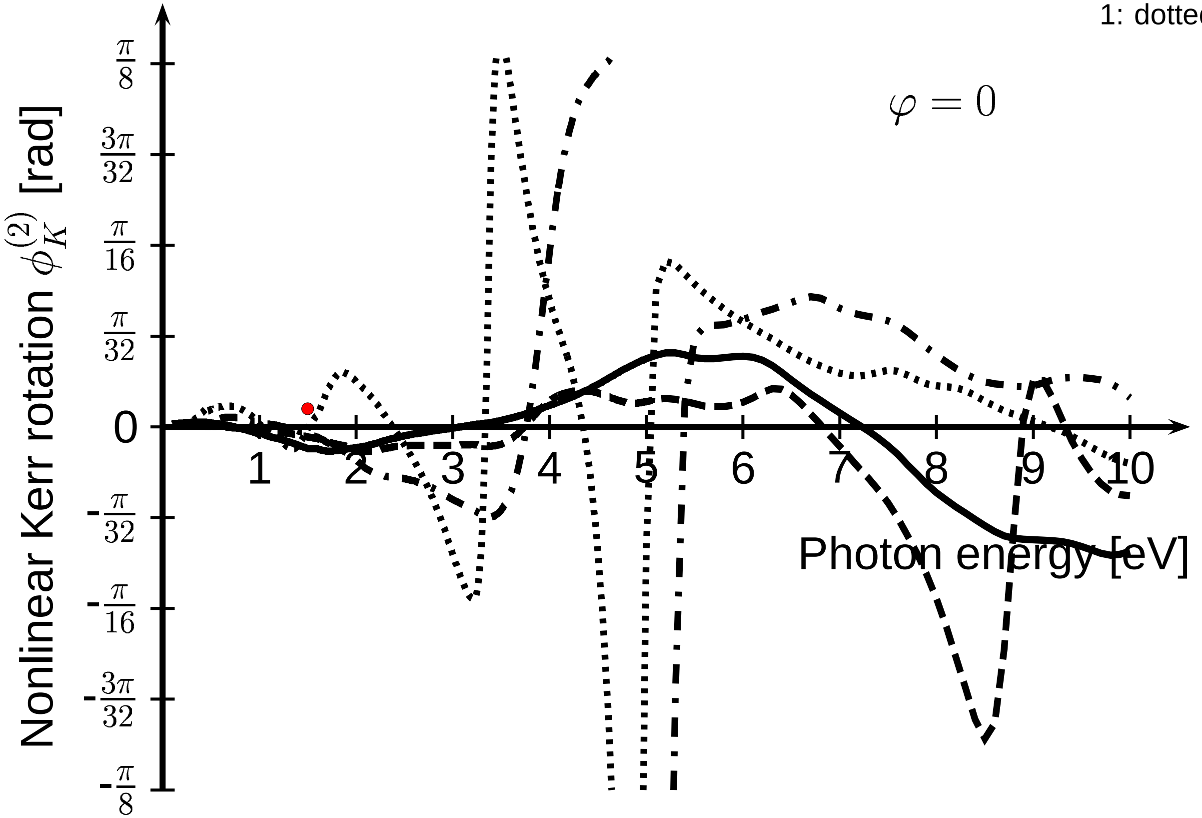
Cu layers
4: solid line
3: dashed line
2: dash-dotted line
1: dotted line

Nonlinear (2ω) magneto-optical Kerr rotation

$$\phi^{(2)} = \frac{1}{2}\arctan\frac{2\mathrm{Re}[E_s(2\omega)/E_p(2\omega)]}{1-|E_s(2\omega)/E_p(2\omega)|^2}+\phi_0$$

Cu layers

4: solid line
3: dashed line
2: dash-dotted line
1: dotted line



Conclusions

- large magnetic tensor element – powerful tool
- adding Cu gives another degree of freedom
- nonlinear optics can be calculated using first principles methods (WIEN97)
- symmetry breaking by magnetism can be resolved
- susceptibilities for 3 and 4 Cu underlayers close
- intensities do not look that converged for 4 Cu layers
- Kerr rotation not converged

Further details in: Torsten Andersen and W. Hübner, *Substrate effects in magneto-optical second-harmonic generation from first principles: Fe/Cu(001)*, to appear in Phys. Rev. B.

Outlook

- Wien2k spin-orbit?
- correct occupied d-band and excited state positions – GW
- nonspherical contributions to spin-orbit interaction
- nonlocality ($|\mathbf{q}| \neq 0$)
- removal of translational invariance along z

Nonlocal theory of nonlinear magneto-optics

Mesoscopic systems

- Finite size
- Surfaces
- Interfaces
- Short light pulses

Magnetism

- Spin-orbit coupling
- Relativistic electrons

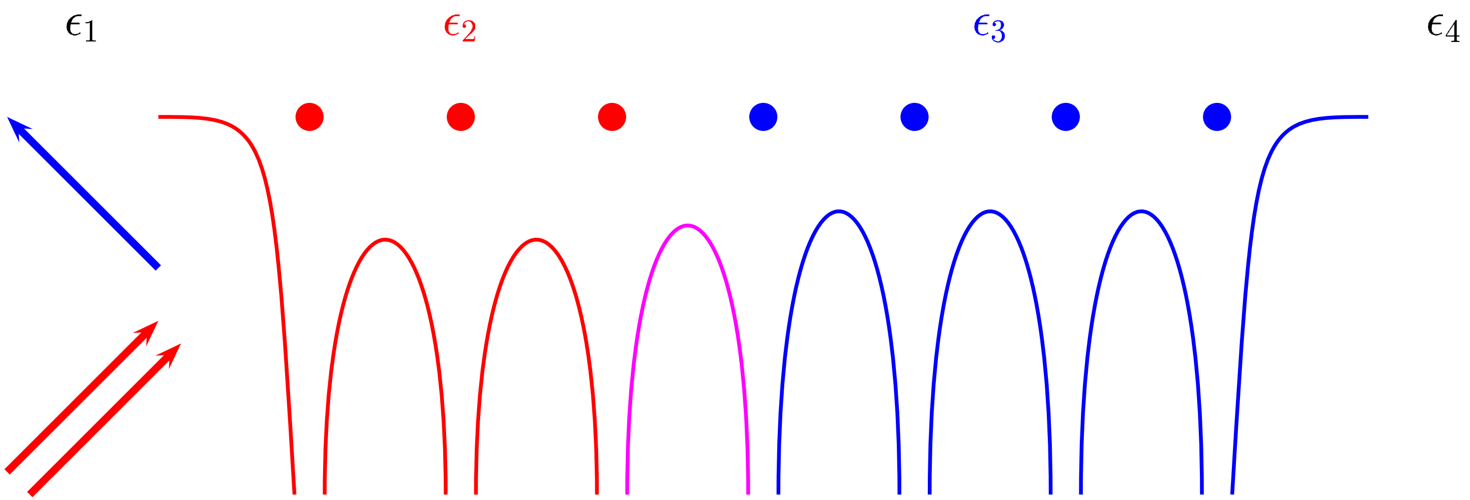
Nonlocality

- Delocalized s-electrons
- Relativistic d-electrons

Problems

- Boundaries?
- Large field gradients $\Rightarrow$ ~~ED~~
- ED $+$ EQ $+$ MD $+ \cdots$?

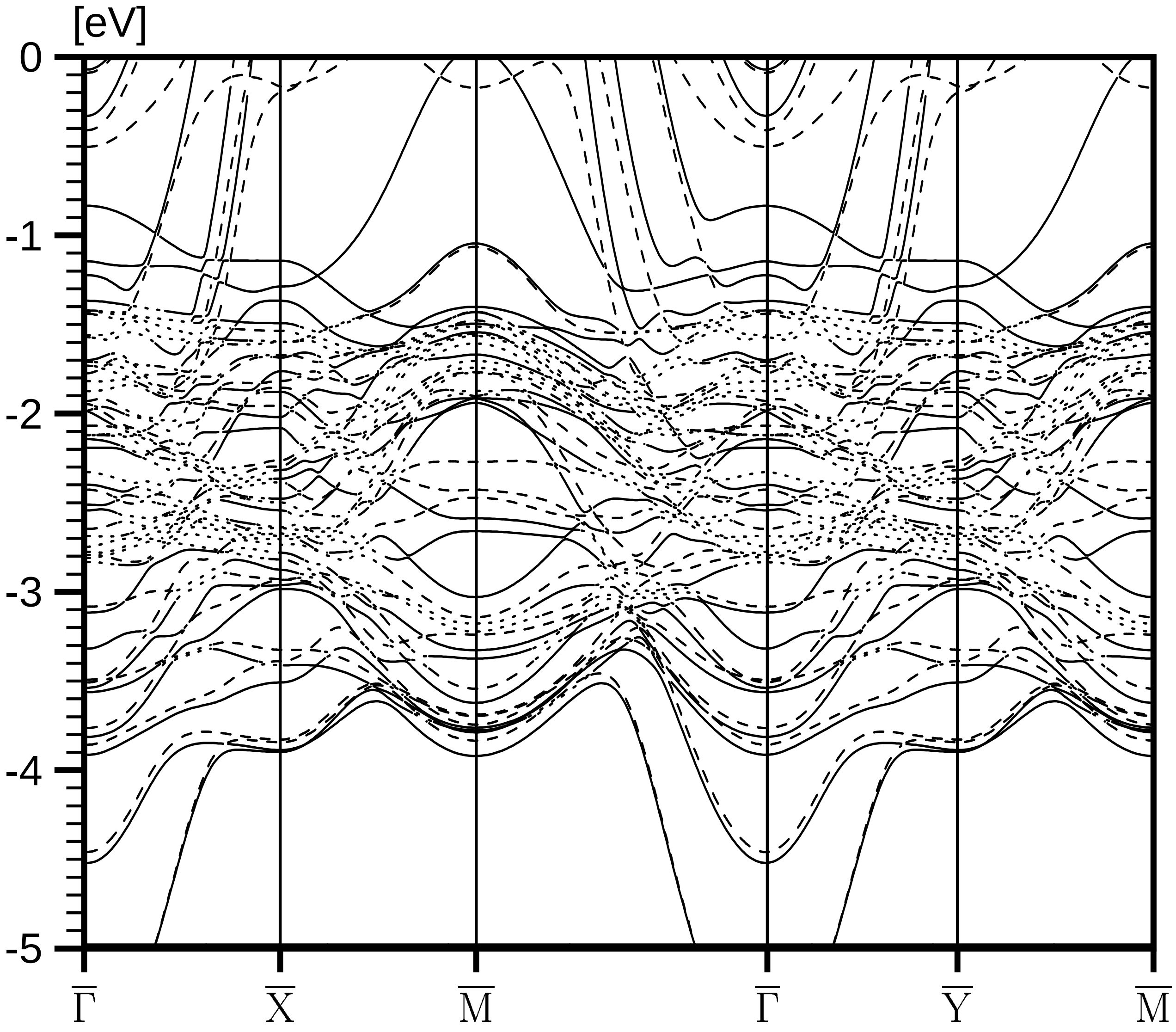
Nonlinear optics



μΦ

Band structure with spin-orbit coupling

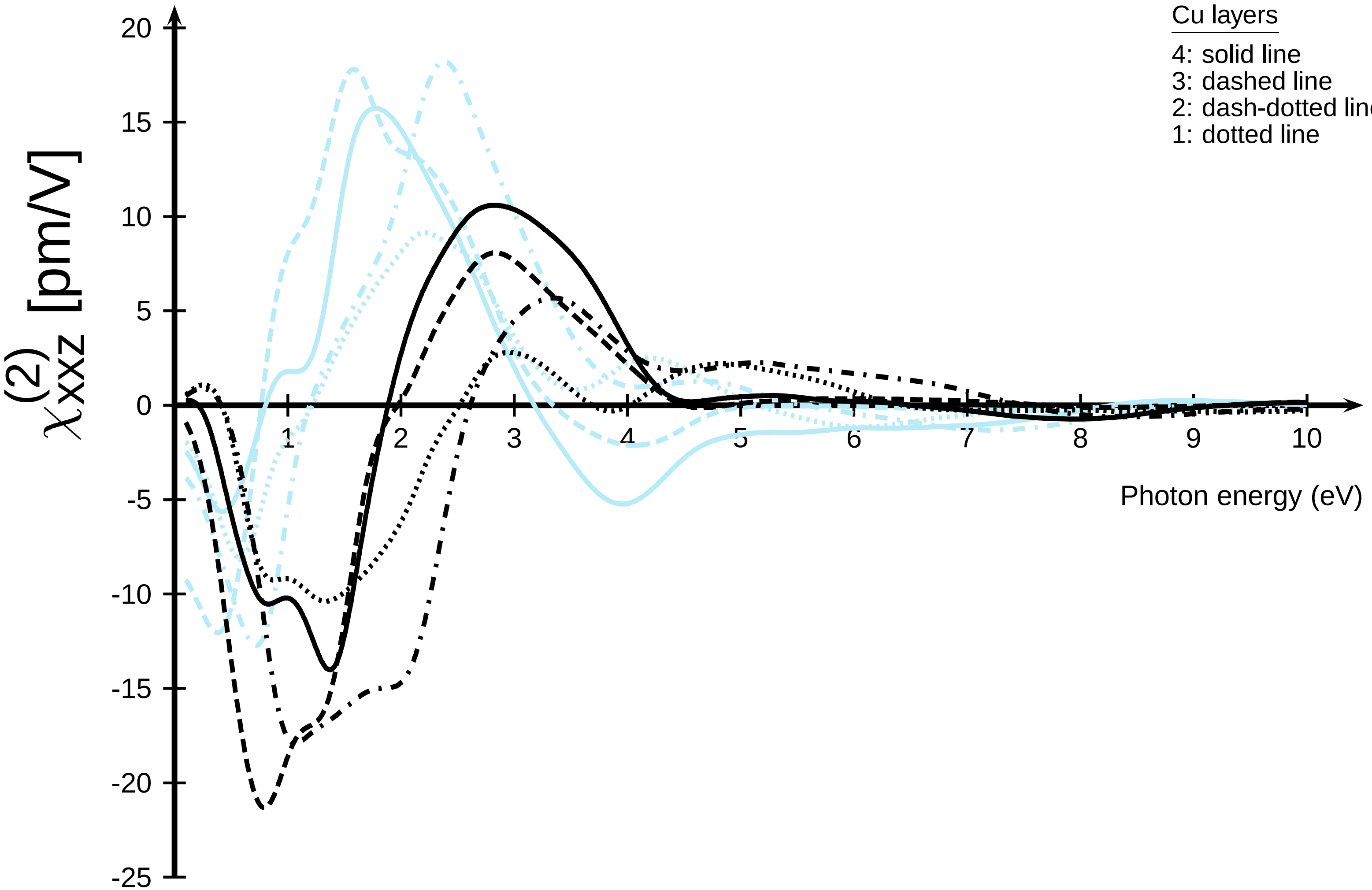
1 ML Fe on 4 ML Cu (001)



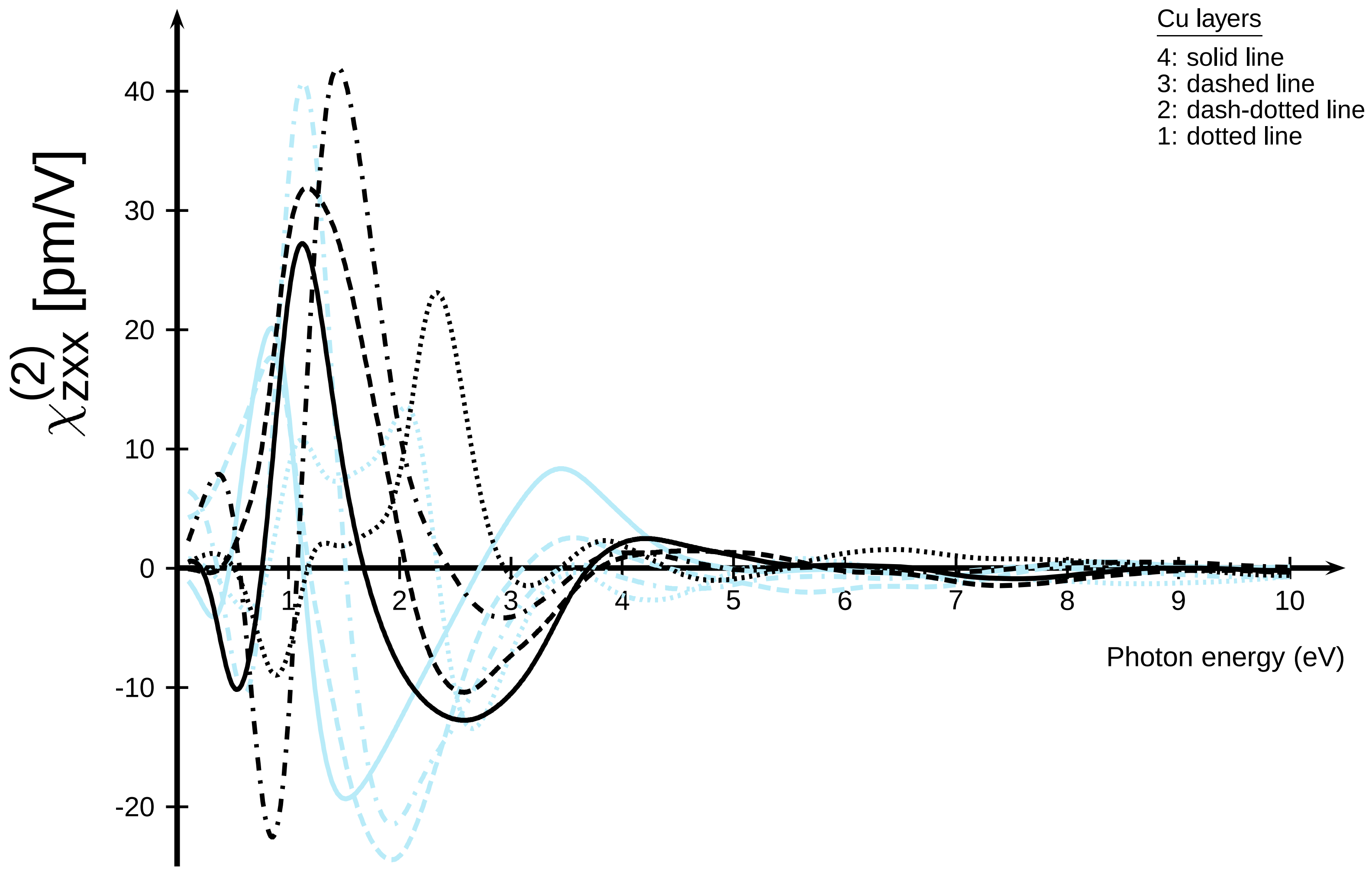
$\mu\Phi$

X2

Screened nonlinear susceptibility tensor



Screened nonlinear susceptibility tensor



Magneto-optical asymmetry $\mathcal{A}$ as function of φ

$$\mathcal{A} = \frac{I(\mathbf{M}\|\mathbf{z}) - I(\mathbf{M}\|-\mathbf{z})}{I(\mathbf{M}\|\mathbf{z}) + I(\mathbf{M}\|-\mathbf{z})}$$

$\hbar\omega$=1.5 eV

Cu layers

4: solid line
3: dashed line
2: dash-dotted line
1: dotted line