Elastic scattering of x-rays by atoms

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Together with:

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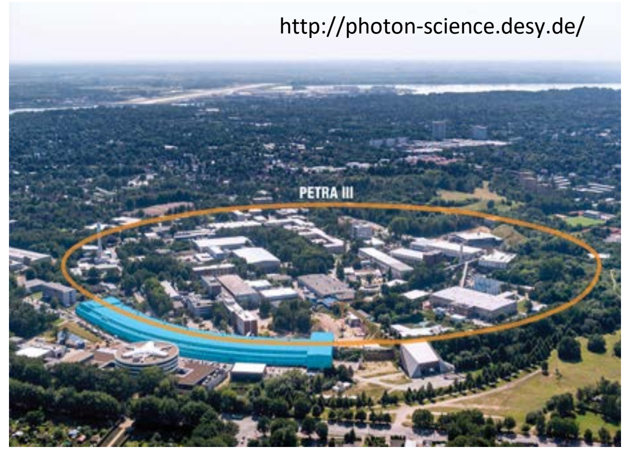
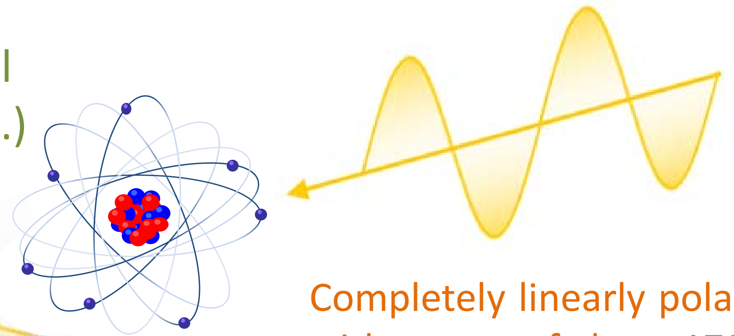
Elastic photon scattering: New experiments

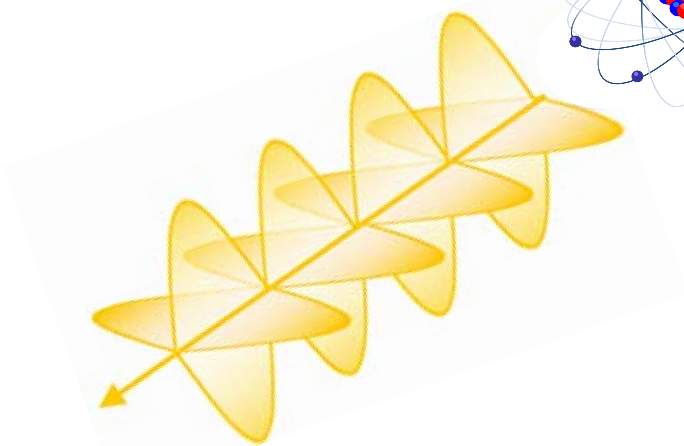
- In the last years, experiments have been performed at the PETRA III facility in DESY to study the elastic x-ray scattering by heavy neutral atoms.



Heavy neutral atom (Au, Pb…)



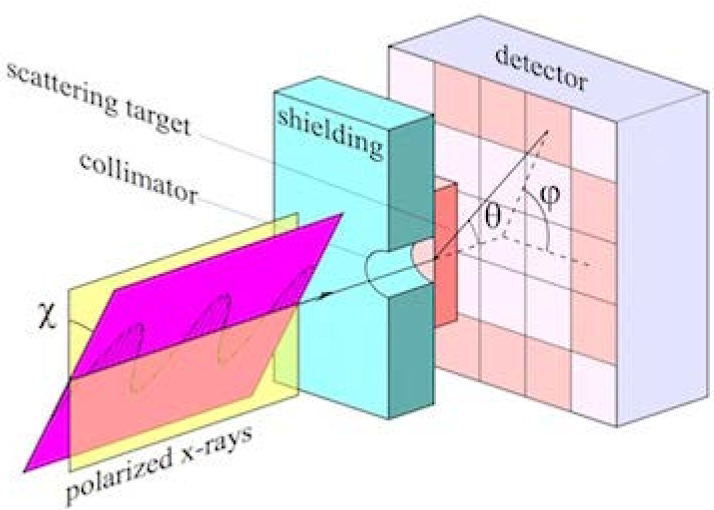
Completely linearly polarized light with energy of about 170 keV



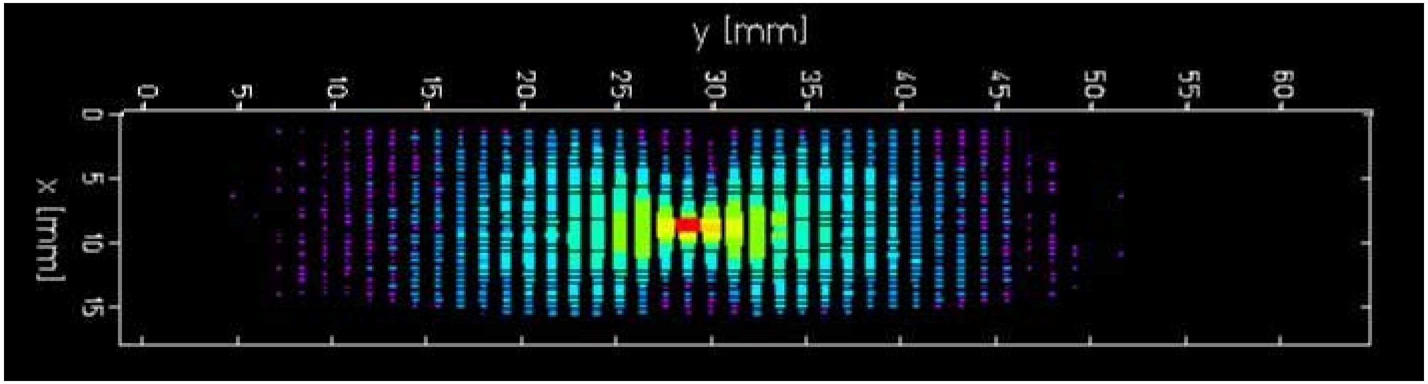
What is the angular distribution and polarization of scattered light?

- Experimentally, the angular and polarization properties of scattered light can be studied with the help of solid-state position-sensitive detectors.

X-ray polarization measurements



A broad range of instruments for x-ray polarimetry for the energy interval up to a several MeV have been developed during the last decade. These instruments use principles of Compton polarimetry.



2D image for Compton scattering of almost 98% linearly polarized x-rays (210 keV) (preliminary result). The image displays the spatial distribution of Compton scattered photons.

Th Stöhlker et al., J. Phys. Conf. Ser. **58**, 411 (2007)

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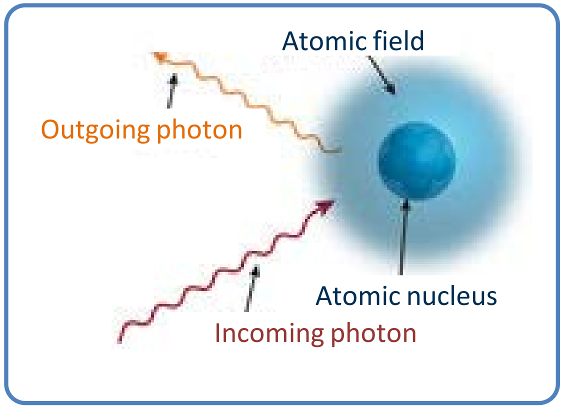
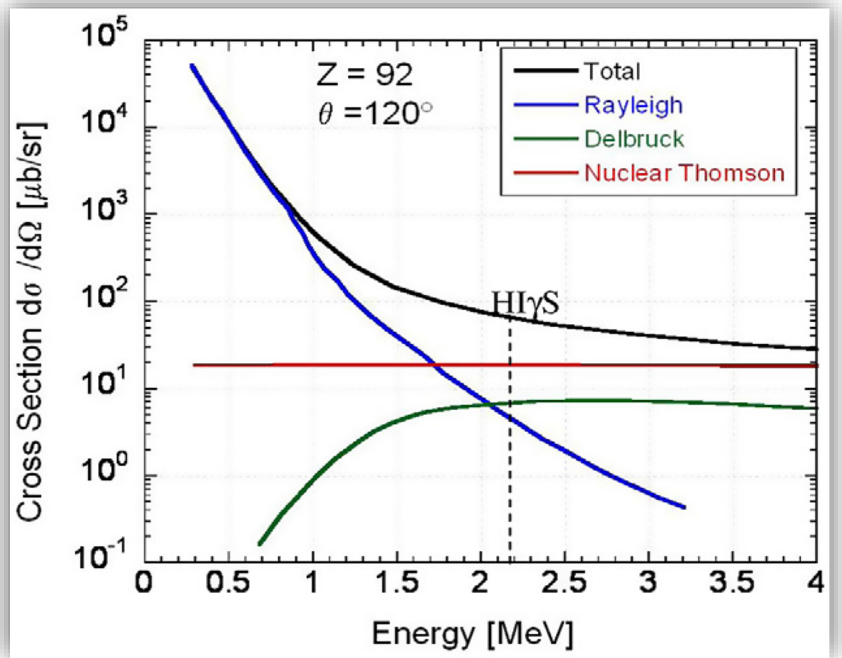
Elastic photon scattering

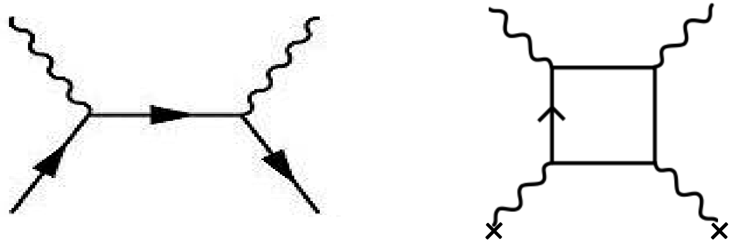
Which processes we refer to when talking about elastic photon scattering?

- Nuclear Thomson scattering (by nucleus)
- **Rayleigh scattering (by bound electrons)**
- Delbrück scattering (by quantum field)





Data from: M.S. Johnson *et al.*, NIMA 285 (2012) 72

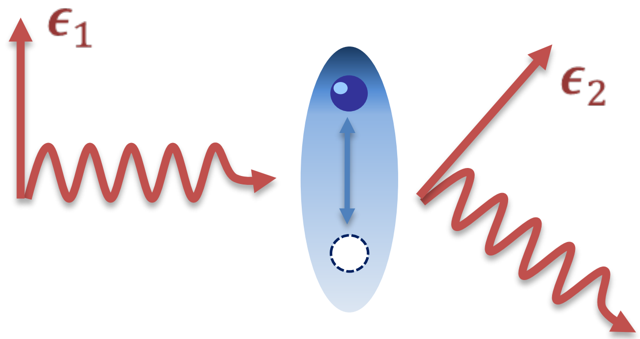


Rayleigh scattering   Delbrück scattering

For incident light with energy of few hundreds keV, the elastic $\gamma + A \rightarrow \gamma + A$ process is dominated by the Rayleigh scattering off bound atomic (or ionic) electrons.

Rayleigh scattering: Naïve approach

What are our naïve expectations about the angular distribution and polarization of scattered radiation for the case when incident photons are completely polarized?



Classical picture: being exposed to linearly polarized light the electron oscillates along the polarization direction and emits dipole radiation:

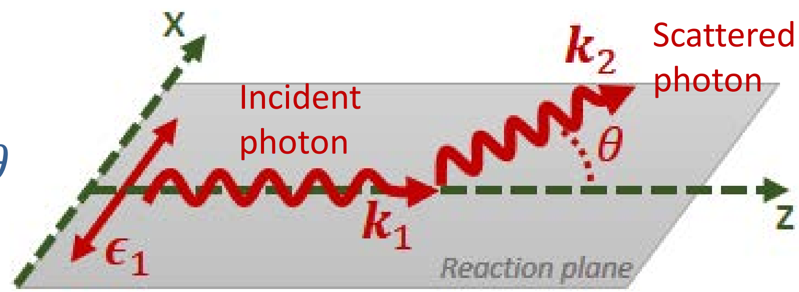
$$\sigma(\boldsymbol{k}_1, \boldsymbol{\epsilon}_1, \boldsymbol{k}_2, \boldsymbol{\epsilon}_2) \propto (\boldsymbol{\epsilon}_1 \boldsymbol{\epsilon}_2^*)^2$$

Polarization vector of incident photon

Polarization vector of outgoing photon

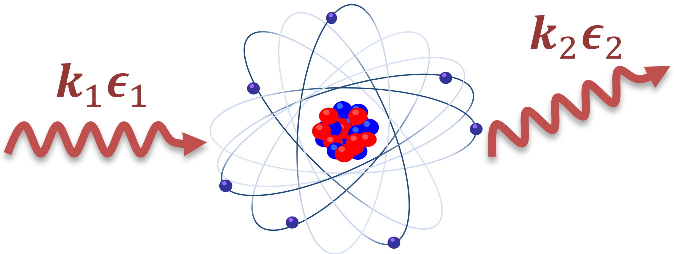
For the *coplanar* case when the photon is scattered in the polarization plane we find:

- angular distribution: $\frac{d\sigma}{d\Omega} = \frac{d\sigma_\parallel}{d\Omega} + \frac{d\sigma_\perp}{d\Omega} \sim \cos^2\theta$
- linear polarization: $P = \frac{d\sigma_\parallel - d\sigma_\perp}{d\sigma_\parallel + d\sigma_\perp} = 1$



$d\sigma_\parallel$ and $d\sigma_\perp$ correspond to $\boldsymbol{\epsilon}_2$ parallel and perpendicular to the reaction plane

Rayleigh scattering: Many-electron theory



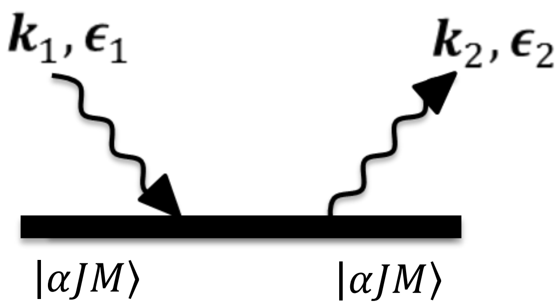
Many-electron atomic state:
$\Psi_{\alpha JM}(\boldsymbol{r}_1, \boldsymbol{r}_2 \ \dots \ \boldsymbol{r}_N)$

For the elastic scattering of low-intensity x-rays by heavy atoms and ions, the electron–photon coupling is described perturbatively. In such an approach, the Rayleigh cross sections are expressed in terms of the second-order transition amplitudes:

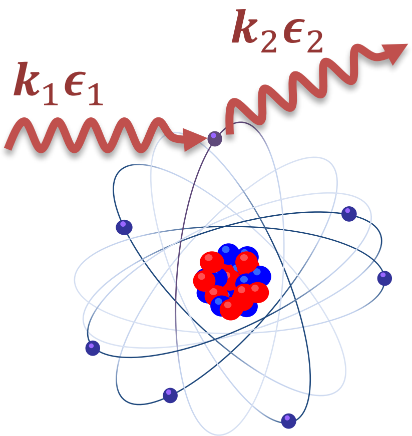
$$M_{at}(\boldsymbol{k}_1\boldsymbol{\epsilon}_1, \boldsymbol{k}_1\boldsymbol{\epsilon}_1) = \sum_{\nu} \frac{\left\langle \Psi_{\alpha JM} \middle| \sum_i \boldsymbol{\alpha}\,\boldsymbol{\epsilon}_2^*\, e^{-i\boldsymbol{k}_2\boldsymbol{r}_i} \middle| \Psi_\nu \right\rangle \left\langle \Psi_\nu \middle| \sum_i \boldsymbol{\alpha}\,\boldsymbol{\epsilon}_1\, e^{i\boldsymbol{k}_1\boldsymbol{r}_i} \middle| \Psi_{\alpha JM} \right\rangle}{E_i - E_\nu + \omega}$$

Evaluation of the second-order amplitude is a very demanding task since it requires summation over <u>complete set</u> of many-electron states.

We need to use some approximate model!



Single active electron approximation (SAE)



Single active electron approximation implies that the light is scattered by a single (active) electron at a time while the remaining electrons are kept ‘frozen’.

$$M_{at}(\boldsymbol{k}_1\boldsymbol{\epsilon}_1, \boldsymbol{k}_2\boldsymbol{\epsilon}_2) = \sum_{nj\mu} M_{nj\mu}(\boldsymbol{k}_1\boldsymbol{\epsilon}_1, \boldsymbol{k}_2\boldsymbol{\epsilon}_2)$$

summation over all occupied one-particle states

After summation over all one-particle states one derives the matrix element as:

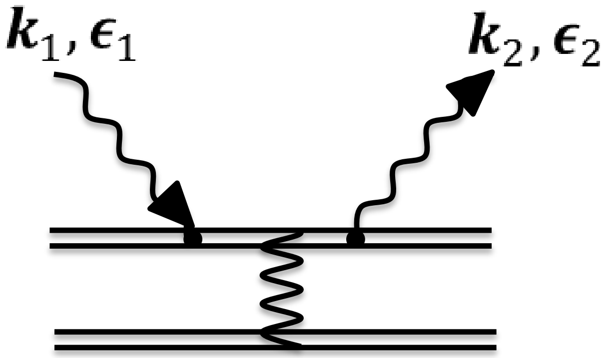
$$M_{at}(\boldsymbol{k}_1\boldsymbol{\epsilon}_1, \boldsymbol{k}_2\boldsymbol{\epsilon}_2) = f(q^2)\,(\boldsymbol{\epsilon}_1 \cdot \boldsymbol{\epsilon}_2^*) + \mathrm{g}(q^2)(\boldsymbol{\epsilon}_1 \cdot \hat{\boldsymbol{n}}_2)(\boldsymbol{\epsilon}_2^* \cdot \hat{\boldsymbol{n}}_1)$$

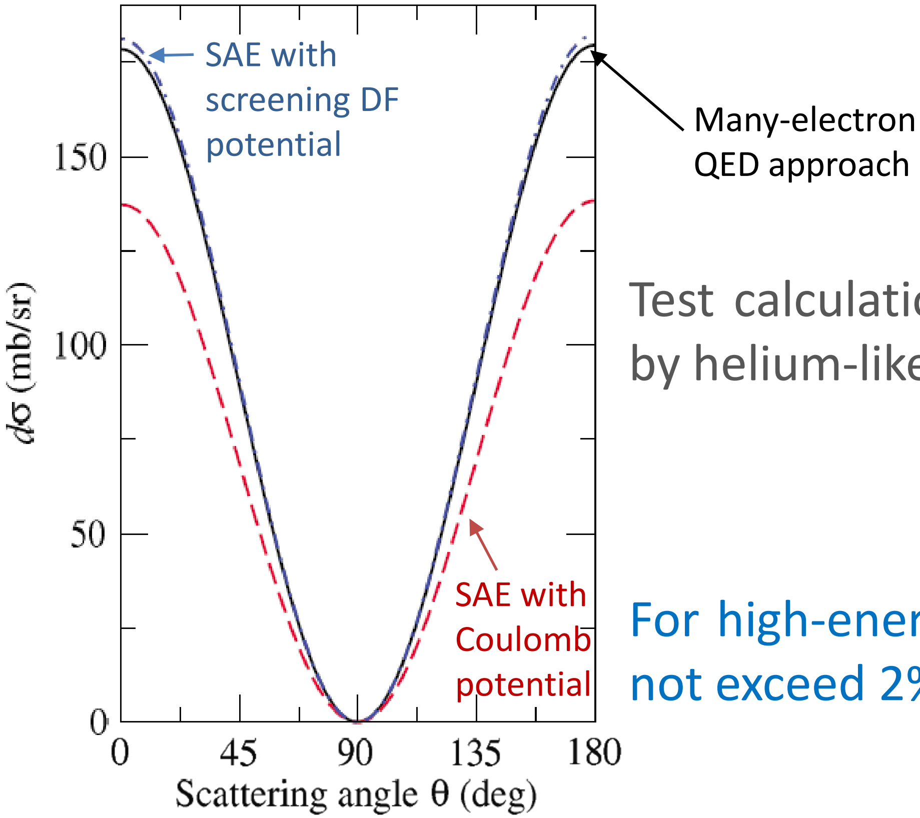
where $\hat{\boldsymbol{n}}_{1,2} = \boldsymbol{k}_{1,2}/k_{1,2}$ and f and g are functions of the square of transferred momentum $q^2 = 4k^2 \sin^2\theta$.

This expression is valid for the scattering by an atom (ion) with *J* = 0 and can also be obtained from symmetry reasoning.

Validity of the SAE approach

A many-electron perturbation theory, based on a rigorous quantum electrodynamics (QED) approach, was developed to check the accuracy of the single-active electron approach (SAE).



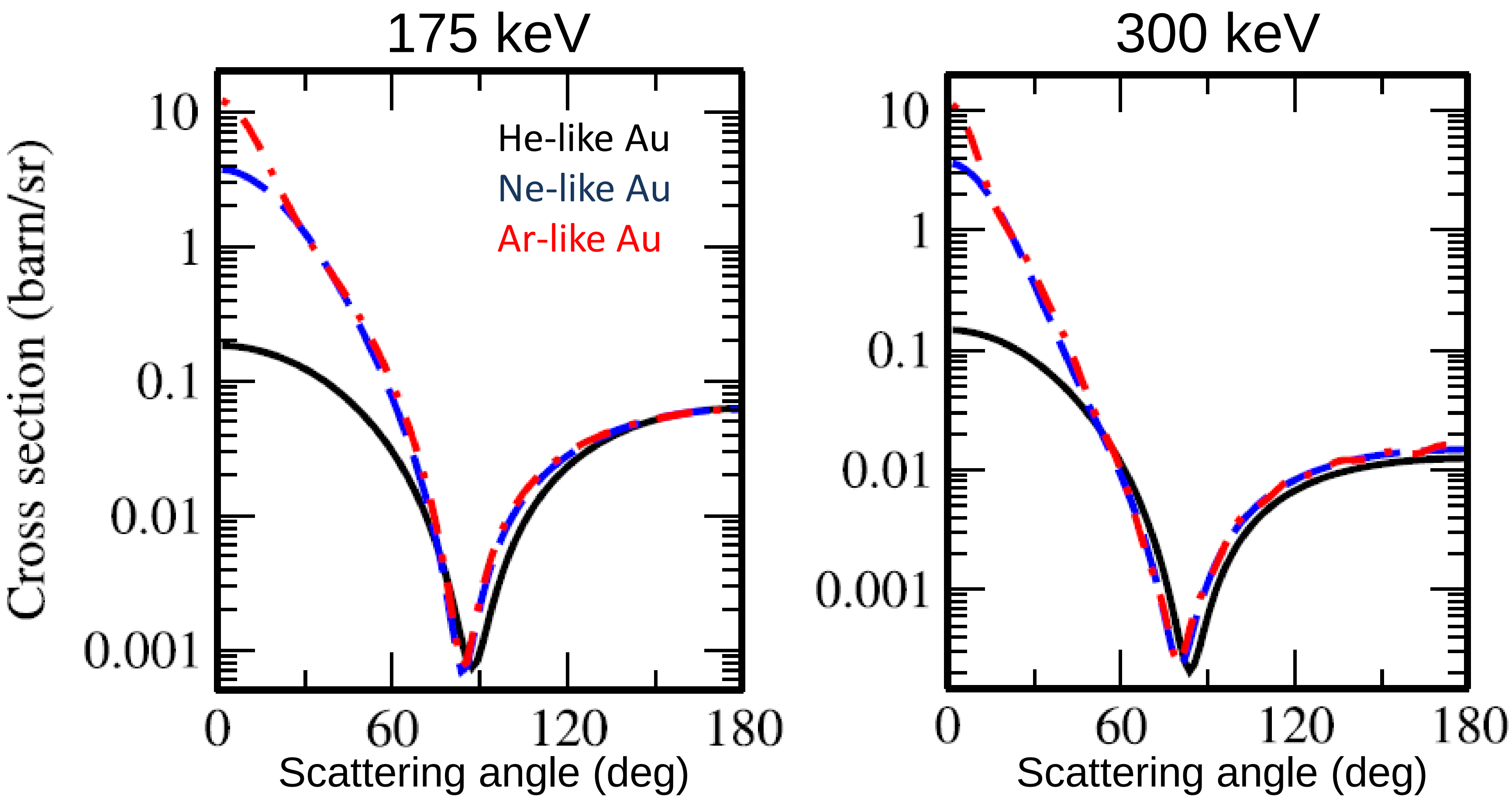


Test calculations: scattering of **polarized** 6-keV photons by helium-like Ni^{26+} ions in their ground state.

For high-energy photons, the effects beyond the IPA do not exceed 2% for the scattering by closed inner shells!

A. Volotka, V. A. Yerokhin, A.S., Th. Stöhlker and S. Fritzsche, PRA **93** (2016) 023418

X-ray scattering by highly-charged ions



The calculations indicate a dramatic change in the angle-differential cross sections with regard to the charge state of the target, while the polarization is insensitive to the shell structure and the number of bound electrons of a particular ion.

A.S., V. A. Yerokhin, Th. Stöhlker and S. Fritzsche, J. Phys. B **48** (2015) 144015

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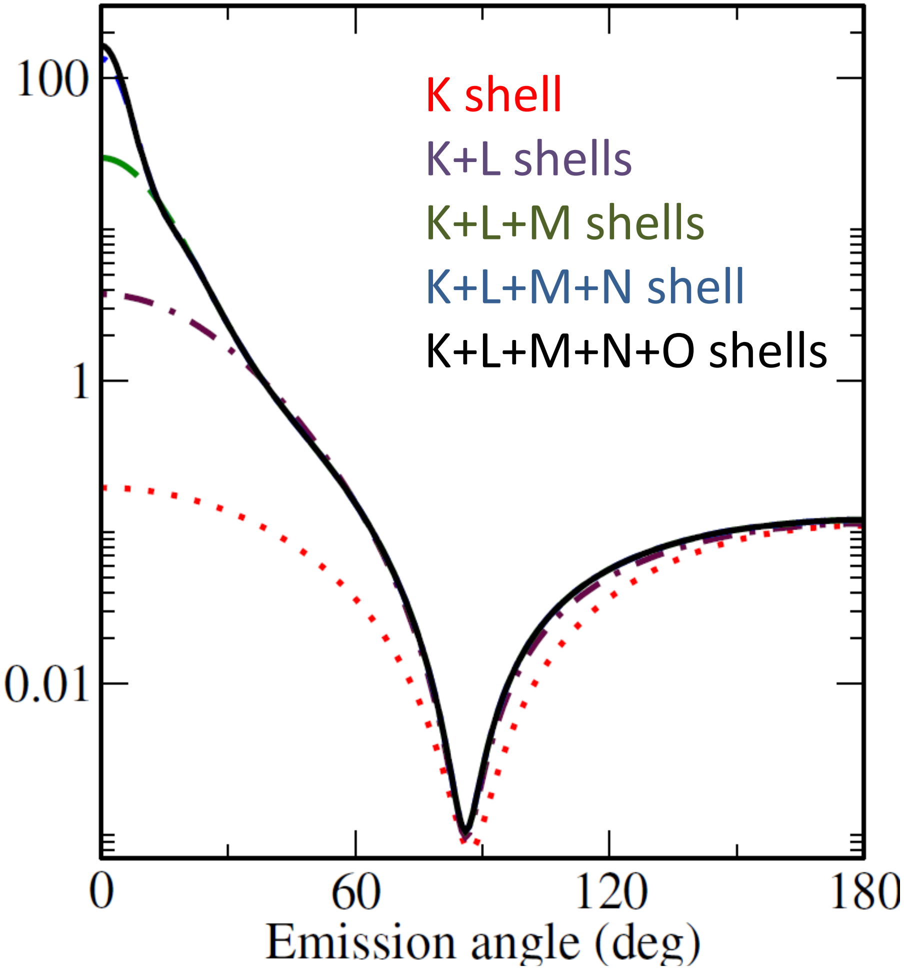
Angular distribution of scattered electrons

Scattering of completely polarized 145 keV photons by Pb atom



Three “scattering regimes” can be identified from our calculations:

- Small scattering angles ($\theta < 30$ deg): small transferred momentum, outer shells play on important role
- Large scattering angles ($\theta > 150$ deg): large transferred momentum, scattering is mainly due to inner K- and L-shells
- Scattering around $\theta = 90$ deg: electric-dipole forbidden region

A.S., V. A. Yerokhin, Th. Stöhlker and S. Fritzsche, J. Phys. B **48** (2015) 144015

Polarization of scattered electrons

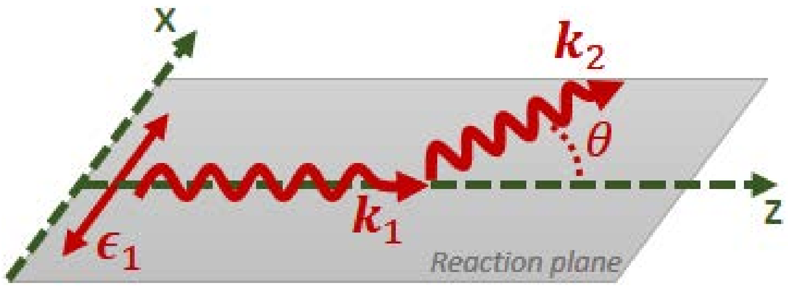
In contrast to the angular distribution, the polarization of scattered photons is rather "boring" if incident light is itself polarized:

$$P = \frac{d\sigma_{\parallel} - d\sigma_{\perp}}{d\sigma_{\parallel} + d\sigma_{\perp}} = 1$$

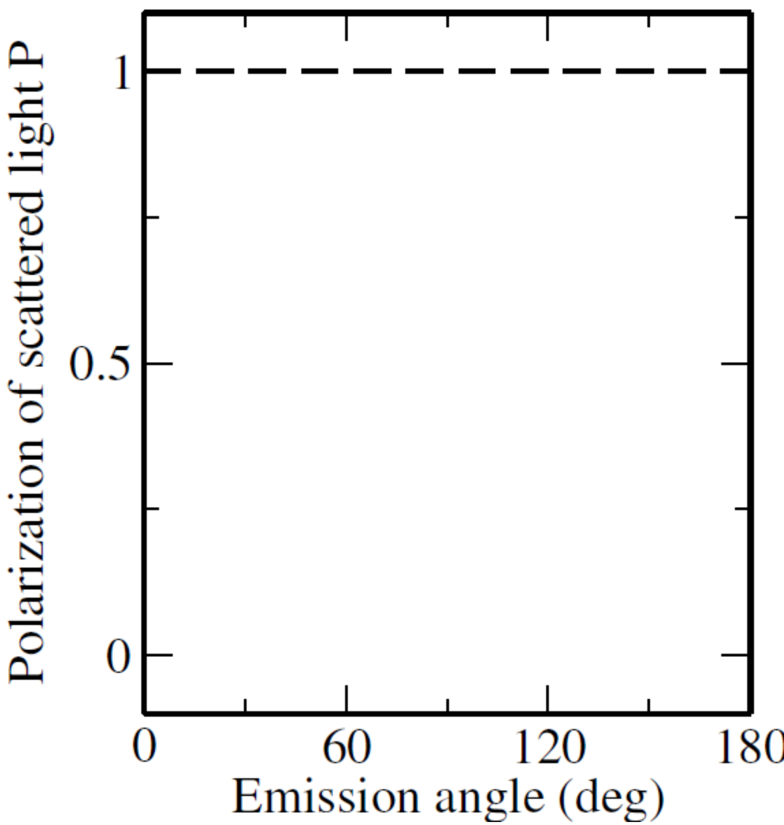
This can be understood from the symmetry properties of the matrix element:

$$M_{at} = f(q^2)\,(\boldsymbol{\epsilon}_1 \cdot \boldsymbol{\epsilon}_2^*) + g(q^2)(\boldsymbol{\epsilon}_1 \cdot \hat{\boldsymbol{n}}_2)(\boldsymbol{\epsilon}_2^* \cdot \hat{\boldsymbol{n}}_1)$$

For the coplanar geometry (our case!): $M_{at} = 0$ if $\boldsymbol{\epsilon}_2$ is normal to reaction plane and, hence $d\sigma_{\perp} = |M_{at}|^2 = 0$



Scattering by Pb atom in its ground state: [Xe] $4f^{14}$ $5d^{10}$ $6s^2$ $6p^2$: J =0



Comparison with experimental data

The Rayleigh scattering of 175 keV photons by neutral gold atoms ([Xe] $4f^{14}$ $5d^{10}$ $6s^{1}$) has been measured by the group of Prof. Stöhlker at the PETRA III facility in DESY.

What could be the reason for such a disagreement?

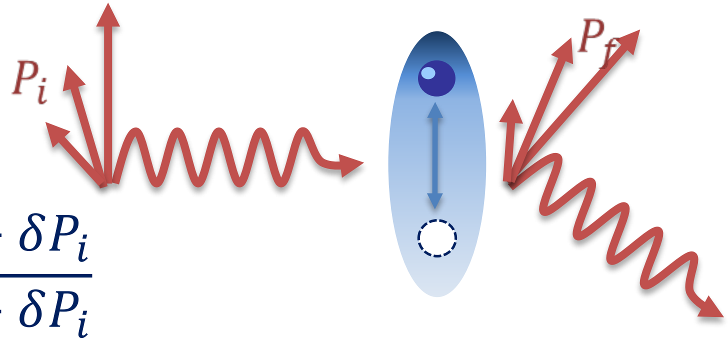
- Outer-shell effects? But we believe that we can reasonably describe them. Moreover, they should be important for the forward emission.
- Polarization effect of the incident light? But we believe that synchrotron radiation is strongly (almost 100 %) polarized. However…

A good agreement between experiment and theory was found for the angle-differential cross section. But not for polarization of scattered light…

K.-H. Blumenhagen *et al.*, New J. Phys. **18** (2016) 103034

Polarization transfer in photon scattering

In order to analyze the polarization effects in the Rayleigh scattering, we have again considered the non-relativistic dipole case:



$$P_f = \frac{(\cos^2\theta - 1) + (\cos^2\theta + 1)P_i}{(\cos^2\theta + 1) + (\cos^2\theta - 1)P_i} \cong \frac{2\cos^2\theta - \delta P_i}{2\cos^2\theta + \delta P_i}$$

Polarization of scattered light

Polarization of incident light

And assumed that the polarization of incident radiation is very high, but not exactly 100%:

$$P_i = 1 - \delta P_i, \quad \delta P_i \ll 1$$

For the forward scattering angles:

$$P_f \approx 1 - \delta P_i$$

The polarization of scattered light is linearly dependent on the "depolarization" parameter and is unity for $\delta P_i = 0$.

For the scattering angles $\theta \approx 90$ deg:

$$P_f \approx -1 + \frac{4\cos^2\theta}{\delta P_i}$$

The polarization of scattered light is inversely proportional to "depolarization"! Small change of δP_i leads to a huge change of P_f.

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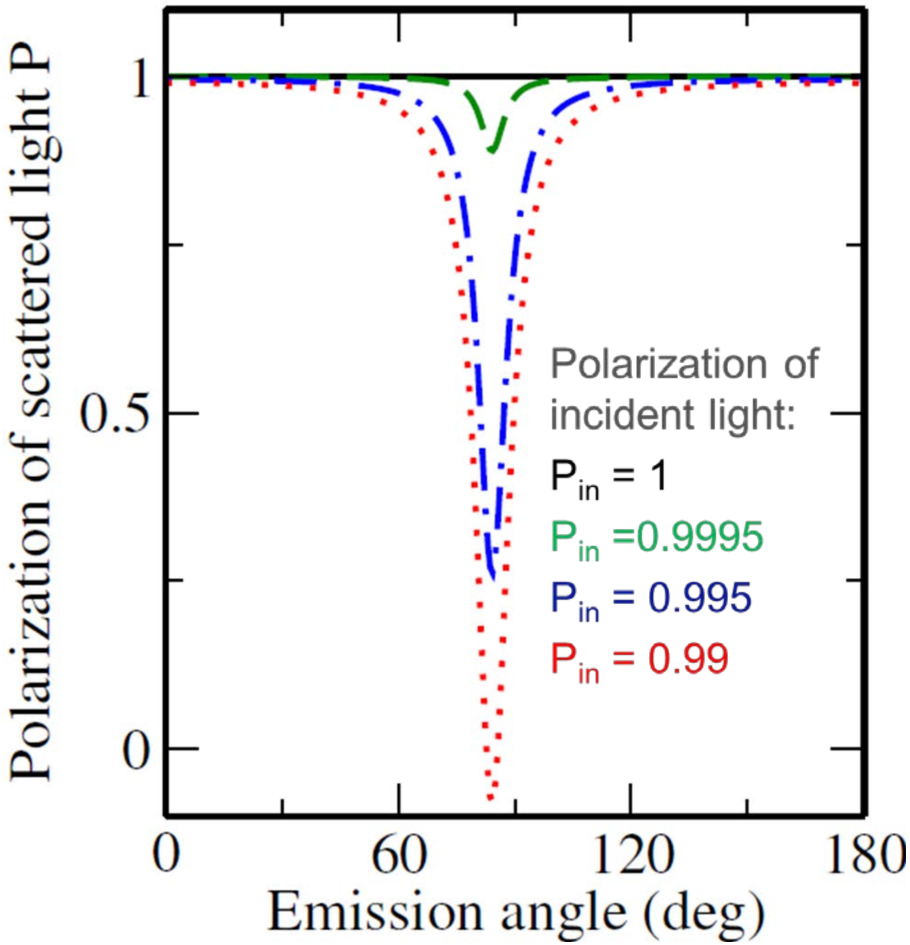
Polarization transfer in photon scattering

Results of calculations for the scattering of 175 keV photons by gold atoms.



The non-relativistic dipole prediction for the scattering angles $\theta \approx 90$ deg is:

$$P_f \approx -1 + \frac{4\cos^2\theta}{\delta P_i}$$

This result has been qualitatively confirmed by the SAE relativistic theory.

This implies that the Rayleigh scattering can be used as a tool for measuring the polarization purity of syncrotron radiation.

K.-H. Blumenhagen *et al.*, New J. Phys. **18** (2016) 103034

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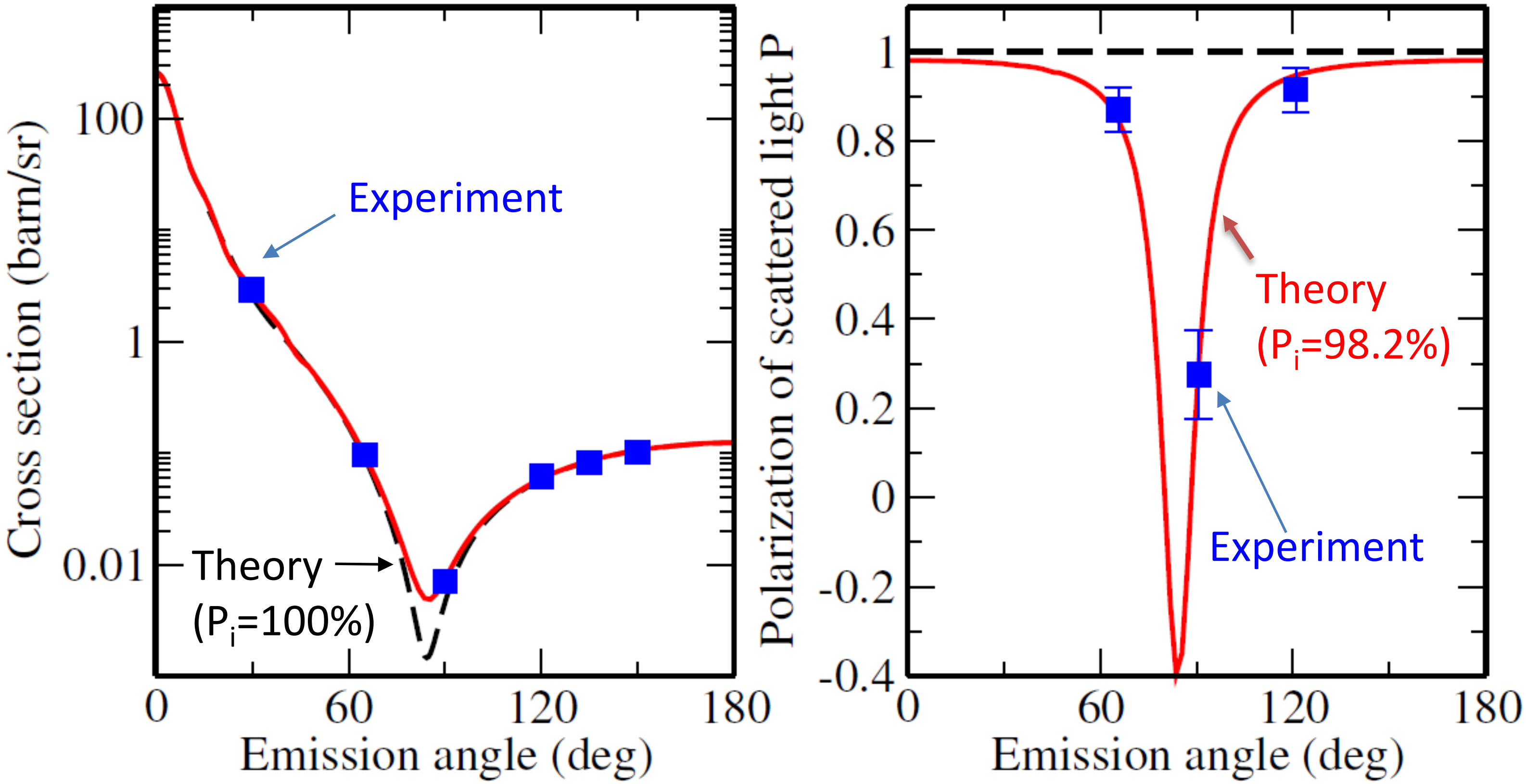
Comparison with experimental data

The Rayleigh scattering of 175 keV photons by neutral gold atoms has been measured by the group of Prof. Stöhlker at the PETRA III facility in DESY.



Based on the comparison between experiment and theory we were able to determine polarization of PETRA III beam as 98.24 ± 0.85 %.

K.-H. Blumenhagen *et al*., New J. Phys. **18** (2016) 103034

This is nice but we want to more efficient method for polarization analysis as just running of tens of numerical calculations.

Analysis of scattering amplitude

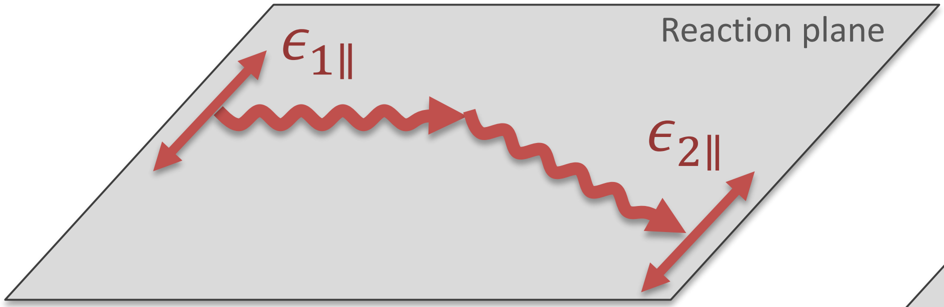
For the scattering by closed-shell systems (J=0) the transition amplitude can be derived from general symmetry analysis:

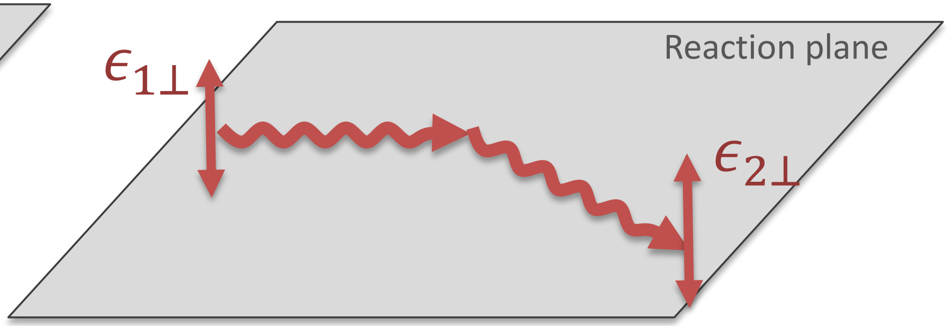
$$M_{at}(\boldsymbol{k}_1\boldsymbol{\epsilon}_1, \boldsymbol{k}_2\boldsymbol{\epsilon}_2) = f(q^2)\,(\boldsymbol{\epsilon}_1 \cdot \boldsymbol{\epsilon}_2) + g(q^2)(\boldsymbol{\epsilon}_1^* \cdot \hat{\boldsymbol{n}}_2)(\boldsymbol{\epsilon}_2^* \cdot \hat{\boldsymbol{n}}_1)$$

For our analysis it is more convenient to re-write it in the form:

$$M_{at}(\boldsymbol{k}_1\boldsymbol{\epsilon}_1, \boldsymbol{k}_2\boldsymbol{\epsilon}_2) = \mathcal{A}_{\parallel}\epsilon_{1\parallel}\epsilon_{2\parallel}^* + \mathcal{A}_{\perp}\epsilon_{1\perp}\epsilon_{2\perp}^*$$

Amplitude is given as two terms, describing scattering photons, polarized within and perpendicular to the reaction plane.





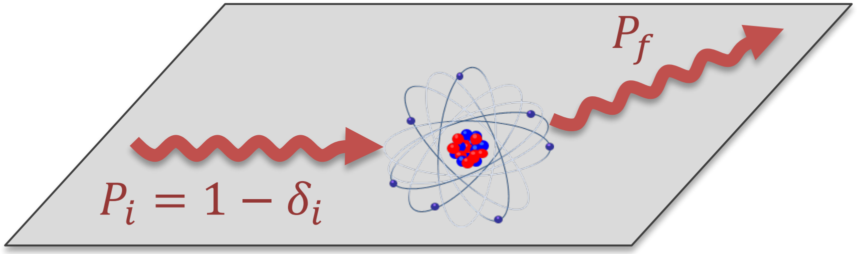
Polarization of scattered light

Just a reminder: we describe the (degree of) polarization of light by the Stokes parameter:

$$P = \frac{I_\parallel - I_\perp}{I_\parallel + I_\perp}$$

Polarization of the scattered light depends on the depolarization $\delta_i = 1 - P_i$ of incident light:

$$P_f(\theta) = 1 - \frac{2}{1 + \frac{2 - \delta_i}{\delta_i} R(\theta)}$$



… and on the ratio of squares of scattering amplitudes: $R(\theta) = \frac{|\mathcal{A}_\parallel|^2}{|\mathcal{A}_\perp|^2}$

This expression is exact for *any* closed-shell target!

But… we need to know $R(\theta)$!

A. S., V. A. Yerokhin, S. Fritzsche, and A. V. Volotka, Phys. Rev. A 98 (2018) 053403

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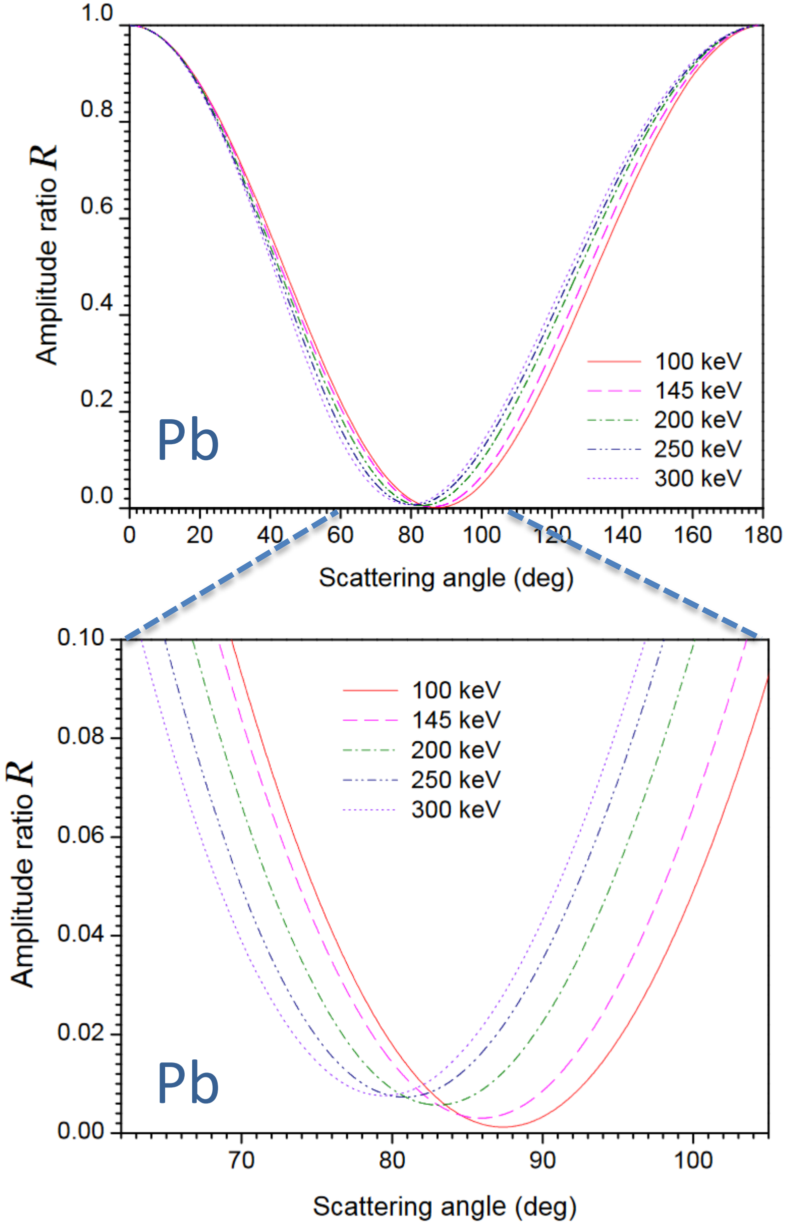
Amplitude ratio $R(\theta)$



The amplitudes can be calculated within the so-called form-factor (FF) approximation, in which the photon scatters off a static charge distribution of individual electrons as obtained from the Dirac-Fock equation:

$$\mathcal{A}_{\parallel} = -\mathcal{F}_n(q)\cos\theta$$

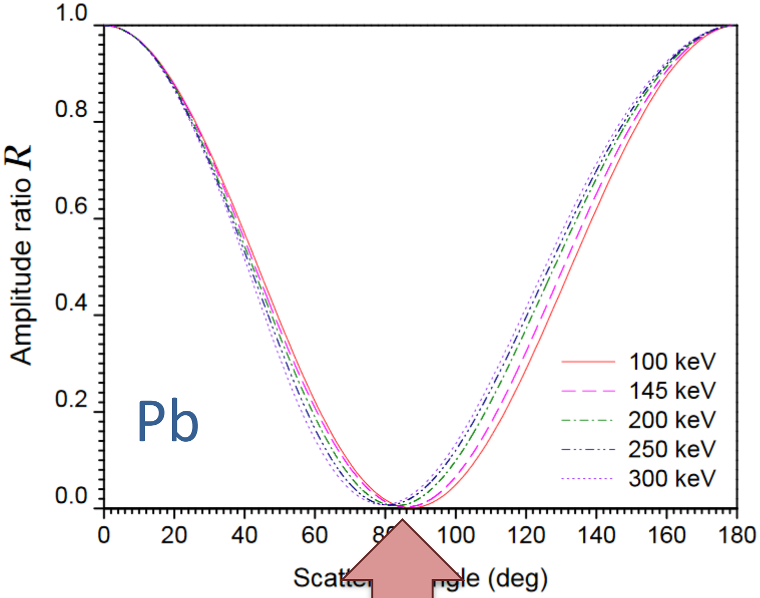
$$\mathcal{A}_{\perp} = -\mathcal{F}_n(q)$$

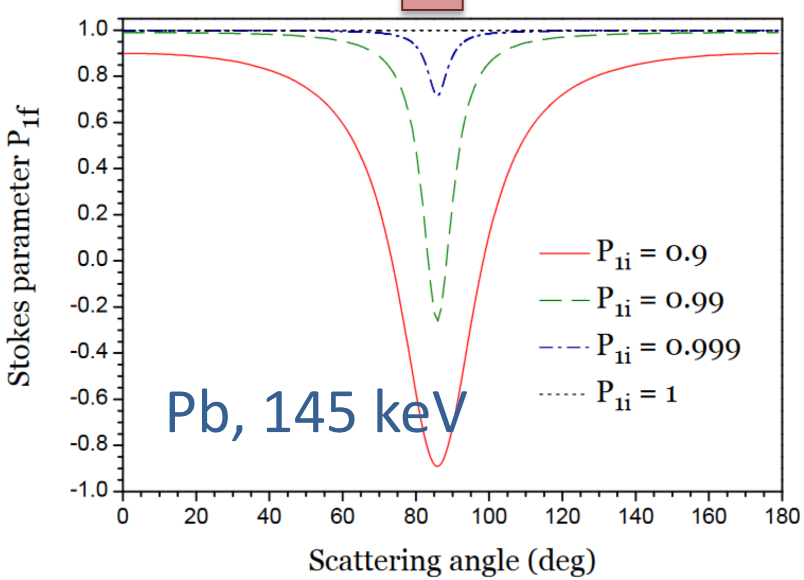
$$R(\theta) = \cos^2\theta$$

“Exact” calculations show deviation from this simple formula.

A. S., V. A. Yerokhin, S. Fritzsche, and A. V. Volotka, Phys. Rev. A 98 (2018) 053403

Polarization purity analysis





$$P_f(\theta) = 1 - \frac{2}{1 - R(\theta) + 2\frac{R(\theta)}{\delta_i}}$$

By measuring the Stokes parameter of the outgoing radiation P_{1f} in the **region of small values of $R(\theta)$**, we achieve an enhanced sensitivity of our measurement to small depolarizations of the incoming radiation.

The theory can:

- Suggest the best angle for polarization purity measurements
- Extract polarization purity from P_{1f}

Calculations of the ratio $R(\theta)$

TABLE II: The ratio of the squares of the parallel and perpendicular amplitudes $R(E,\theta)$ for Rayleigh scattering off the neutral lead atom. "FF" denotes the (standard) form-factor approximation, with $R_{\mathrm{FF}}(\theta) = \cos^2\theta$.

θ (deg)	E (keV)	R
30	FF	0.7500
	100	0.7394 (3)
	145	0.7335 (3)
	200	0.7224 (4)
	250	0.7148 (10)
	300	0.7095 (15)
60	FF	0.2500
	100	0.2212 (4)
	145	0.2077 (6)
	200	0.1888 (7)
	250	0.1646 (9)
	300	0.1425 (15)
70	FF	0.1170
	100	0.0926 (4)
	145	0.0837 (4)
	200	0.0663 (6)
	250	0.0497 (7)
	300	0.0386 (15)
80	FF	0.0302
	100	0.0176 (2)
	145	0.0143 (1)
	200	0.0088 (2)
	250	0.0074 (2)
	300	0.0077 (3)
90	FF	0.0000
	100	0.0030 (1)
	145	0.0085 (1)
	200	0.0225 (2)
	250	0.0352 (8)
	300	0.0436 (18)

$$P_f(\theta) = 1 - \frac{2}{1 + \dfrac{2-\delta_i}{\delta_i} R(\theta)}$$

Detailed calculations of the ratio $R(\theta)$ have been performed for various elements and large range of photon energies.

The uncertainty ascribed to the numerical values of $R(\theta)$ reflects our estimation of the error caused by an incomplete treatment of the outer electron shells.

A. S., V. A. Yerokhin, S. Fritzsche, and A. V. Volotka, Phys. Rev. A 98 (2018) 053403

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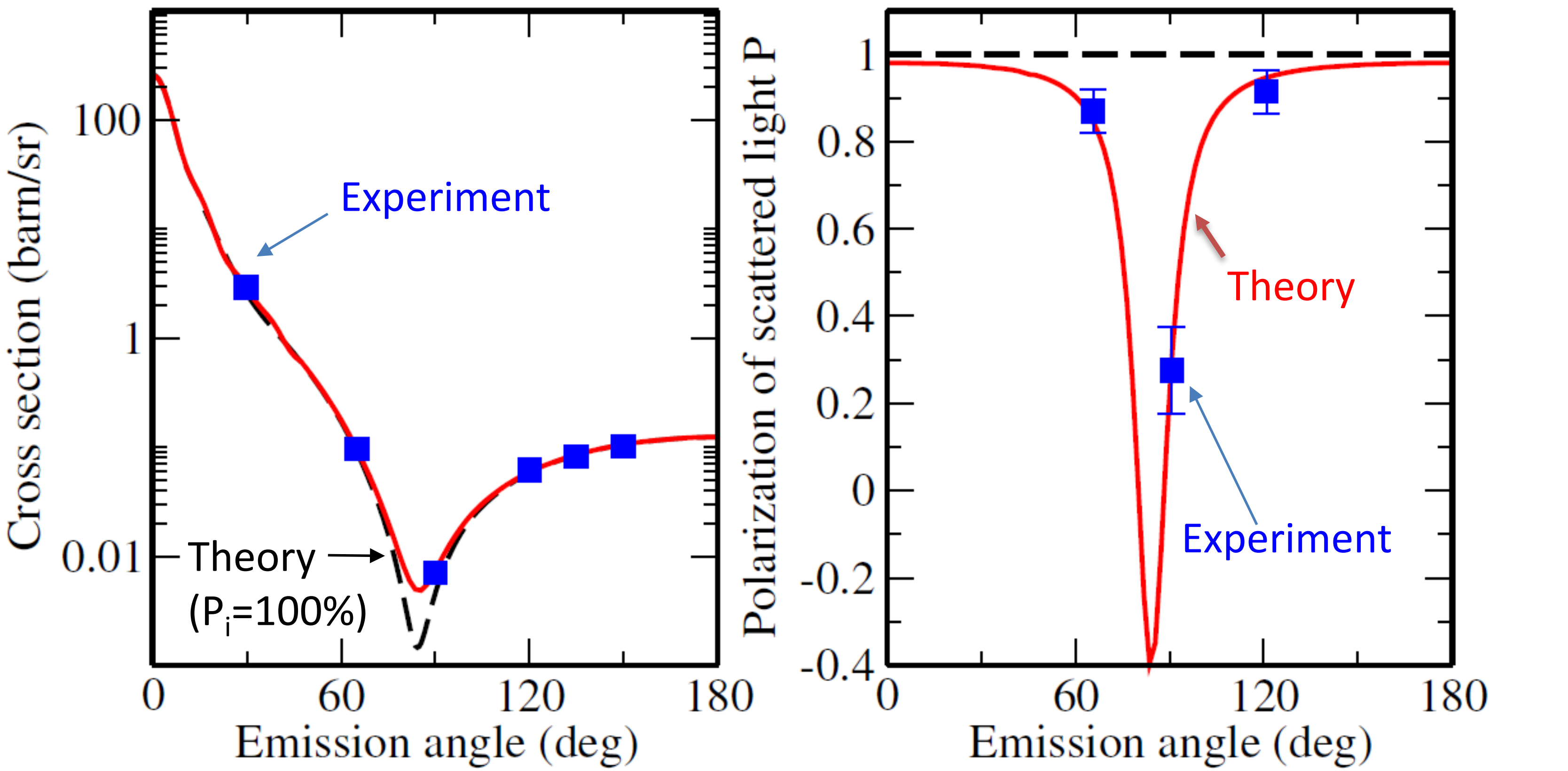
One more time about PETRA III experiment



From the analysis of the Stokes parameter of scattered light at three angles, 60, 90 and 120 degrees, and based on our calculations we predict: $P_i = 98.25 \ \pm 0.72\ \%$.

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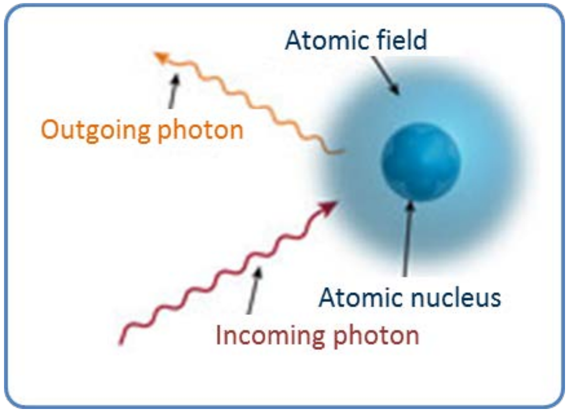
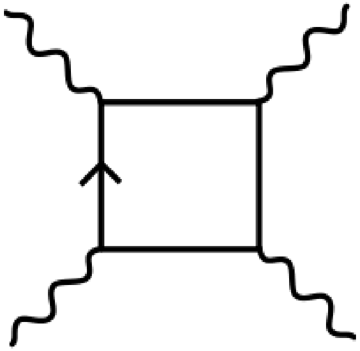
Summary and outlook

- We studied theoretically angular and polarization properties of x-rays elastically scattered by heavy atoms and ions.



- Based on our theoretical analysis we argue that the elastic Rayleigh scattering:
  - Allows us to "probe" both inner- and outer-shell electron structure
  - Provides a new method to measure the polarization purity of x-rays



- For the energies $\hbar\omega \geq 1$ MeV the Delbrück scattering can significantly influence the properties of scattered x-rays.

- Analysis of the Delbrück scattering is our next task.

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