

## Refractive Index of Silicon at $\gamma$ Ray Energies

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For x rays the real part of the refractive index, dominated by Rayleigh scattering, is negative and converges to zero for higher energies. For  $\gamma$  rays a positive component, related to Delbrück scattering, increases with energy and becomes dominating. The deflection of a monochromatic  $\gamma$  beam due to refraction was measured by placing a Si wedge into a flat double crystal spectrometer. Data were obtained in an energy range from 0.18 MeV to 2 MeV. The data are compared to theory, taking into account elastic and inelastic Delbrück scattering as well as recent results on the energy dependence of the pair creation cross section. Probably a new field of  $\gamma$  optics with many new applications opens up.

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In optics the index of refraction  $n(E_\gamma) = 1 + \delta(E_\gamma) + i\beta(E_\gamma)$  is split into a real part  $\delta$  and an imaginary part  $\beta$ , describing refraction and absorption, respectively. In this publication we report on the first measurement of  $\delta$  for silicon up to 2 MeV and the totally unexpected finding that it changes sign above 0.7 MeV, causing  $n$  to be larger than 1. The index deviation  $\delta$  is only  $10^{-9}$  and is explained by Delbrück scattering and virtual pair creation in the high nuclear electric field. We find that higher-order Delbrück scattering gives leading contributions to  $\delta$ . Thus, extrapolating our results towards high  $Z$  atoms like gold, we expect a much larger  $\delta$  in the range of about  $10^{-5}$ , opening up a totally new field of refractive  $\gamma$  optics.

The use of refractive optics in combination with diffracting elements has been a fast developing field for x rays up to 200 keV [1] and has been used for focusing to a small spot size ( $\mu\text{m}$ -10 nm) [2]. The refractive real part can be directly calculated by Kramers-Kronig dispersion relations from the absorptive cross sections. For x ray lenses it is determined by the virtual photo effect (Rayleigh scattering) and follows the law [3]

$$\delta_{\text{photo}} = -2.70 \frac{\lambda^2 \rho Z}{A} 10^{-6} \propto \frac{1}{E_\gamma^2}. \quad (1)$$

Here the photon wavelength  $\lambda$  is measured in  $\text{\AA}$ ,  $\rho$  in  $\text{g}/\text{cm}^3$ ,  $Z$  is the atomic number, and  $A$  is the atomic mass in grams. Obviously,  $\delta_{\text{photo}}$  converges with  $1/E_\gamma^2$  very fast towards zero, limiting significantly the construction of focusing optics at higher energies. A reasonable focal length  $f$  could be obtained by a large number of lenses  $N$ , since  $f = R/(2\delta N)$ , where  $R$  is the radius at the apex of the parabolically shaped, concave lenses. In case of hard x rays several hundreds lenses are used. For MeV  $\gamma$  rays this number  $N$  would become extremely large and absorption will prevent their use. The argumentation is based on the extrapolation of the  $1/E_\gamma^2$  scaling of the virtual photo

effect, for which experimental knowledge is only available up to about 200 keV.

An experiment following the concept of [4], but using  $\gamma$  rays from an in-pile target at the neutron high-flux reactor at Institut Laue-Langevin (ILL) in Grenoble, was carried out. Samples with a total mass of about 10 g and a total surface of about  $30 \text{ cm}^2$  are placed in a thermal neutron flux of  $4 \times 10^{15}/(\text{s cm}^2)$  and yielding emission rates for  $\gamma$  energies of up to  $10^{15} \text{ s}^{-1}$ . For the present experiment we use very intense transitions of  $^{36}\text{Cl}$  and  $^{156,158}\text{Gd}$  (and during a preparation stage, of  $^{168}\text{Er}$ ). The beam from the sample is collimated over a total distance of 17 m with a cross section of  $4 \times 20 \text{ mm}^2$ . Behind the spectrometer, a movable 3 m long collimation system separates the diffracted beam from the direct beam. The detection is done via a calibrated HP-Ge detector. A schematic layout of the experimental principle is shown in Fig. 1.

The spectrometer is equipped with 2.5 mm thick single crystals of silicon, the lattice spacing of which has been indirectly measured with  $\Delta d/d \simeq 5 \times 10^{-8}$  (for details see [5]). The angular acceptance width of the crystals at  $\gamma$  ray energies is in the order of nanoradian. The spectrometer is set in nondispersive geometry, where the second crystal rocks around the parallel position with respect to the first one. In this geometry the first crystal diffracts an energy band defined by the divergence of the incoming beam (a few microradian). In this geometry the instrument does not act as high-resolution monochromator. The line shapes are obtained via summing counts in a 2 keV wide window of the Ge spectrum, taken for each crystal position. This discriminates background  $\gamma$  rays or unwanted diffraction orders via the energy resolution of the detector. The advantage of the nondispersive geometry is its high sensitivity to beam deflection between the crystals induced by a refractive prism (see below). At the same time systematic errors such as those due to Doppler broadening of the

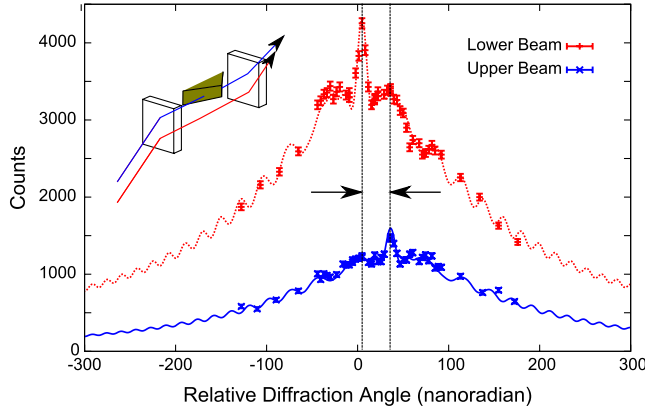


FIG. 1 (color online). Illustration of the measurement principle. The  $\gamma$  beam is coming from the ILL high-flux reactor. A silicon wedge placed between the two crystals of the GAMS5 spectrometer deflects only the upper part of the beam, while the lower beam is propagating in air. The two line shapes were taken during preparation for illustration purposes with the 184 keV line of  $^{168}\text{Er}$  with a very long acquisition time. During the actual measurement, the acquisition time was about 10 times shorter. The upper (blue) beam shows a reduced intensity compared to the lower (red) beam due to absorption in the wedge. The spectra show “pendel solutions” like intensity oscillations typical for a double crystal spectrometer [21]. One clearly sees the angular deflection by the wedge.

$\gamma$  rays (thermal motion, recoil motion) and vertical divergence [6] are at minimum.

Between the two crystals, a silicon prism with an edge angle of  $160^\circ$  and optically polished flat faces was mounted. The upper half of the  $\gamma$  beam was passing through the wedge, while the lower half passed underneath through air. The second crystal is supposed to compare angular deviations of the upper and lower beam with a sensitivity of  $10^{-9}$  rad. The switching between the two halves of the beam is realized via a lead shutter, which is installed on the collimation system behind the spectrometer. This assures that the movement of the lead shutter is mechanically decoupled from the spectrometer. The acquisition time was kept as short as possible to minimize drift problems. For each energy about 30 pairs of scans were taken and averaged. The temperature, pressure, and humidity at the spectrometer were monitored and time variations of the lattice spacing and the refractive index of air were corrected as described in [5]. For background measurements we compared both beams, removing the wedge from the spectrometer. However, this was only carried out once at the end of the measurement, since it represents a serious intervention perturbing the stability of the instrument. The background values were not subtracted from the data, since only a few are available. Further, a direct conversion of the measured deviation angles into absolute values of  $\delta$  is difficult, since it requires an exact knowledge of the absolute alignment of the wedge with respect to the beam. Since we are interested in the energy dependence, we have used the result at 182 keV for

normalization to Eq. (1). The measured values of the index of refraction are compiled in Table I and shown in Fig. 2.

The measurement is based on a homogeneous lattice spacing. The maximum variation of the lattice spacing can be estimated by considering the measured width of the rocking curve and comparing it to calculations of dynamical diffraction theory of ideal crystals. Such evaluations are sensitive for  $\gamma$  ray energies above 700 keV, since the rocking curve is becoming sufficiently narrow. Experimentally, a deviation from theory in the order of 30 nanoradian is observed. It is almost completely related to vibrations of the crystals of about 20 nanoradian, each. A relative variation of the lattice spacing of  $\Delta d/d < 10^{-6}$  could be estimated. A slight energy dependence of the background measurement can be seen. At present we interpret this effect by residual surface tensions on the crystal. In [7] theoretical investigations were carried out indicating that the diffraction angle in Laue geometry is defined within a surface layer. Because the depth of this layer is increasing with increasing energy, the low energy measurements are more sensitive.

An interpretation of the experimental results is possible by demonstrating that the process of virtual pair creation (Delbrück scattering) takes over at higher  $\gamma$  energies with a  $\delta_{\text{pair}}$ , leading to a sign change of the refractive index. In Delbrück scattering the  $\gamma$  ray interacts with the strong electrical field of the nucleus, causes virtual pair creation and then is rescattered. Averaging over a sphere with the diameter of the Compton wave length of the electron (400 fm), we obtain an electric nuclear Coulomb field for silicon of about the Schwinger field of  $E_s = 1.2 \times 10^{18}$  V/m, pointing to a situation where perturbative first-order QED is not sufficient. The dominance of  $\delta_{\text{pair}}$  for higher  $\gamma$  energies can be explained by a refined use of dispersion relations [8–10], which remain valid even for nonperturbative QED.

In the right-hand side of Fig. 3, we show contributions to the absorption cross sections for  $\gamma$  rays [11] together with cross sections of different scattering processes. We divided the important Delbrück scattering into an elastic part, where in a Mössbauer-like recoilless scattering the recoil is taken up by the total crystal  $\sigma_{\text{Delb,elas.}}$  and an inelastic cross section  $\sigma_{\text{Delb,inelas.}}$ , where the scattered  $\gamma$  quantum has lost the recoil energy to the nucleus. We roughly calculate the angle integrated cross sections from theory [12,13] up to the

TABLE I.  $\gamma$  energies with measured index of refraction  $\delta$  for silicon using  $^{36}\text{Cl}$  and  $^{156,158}\text{Gd}$  sources.

$E_\gamma$ keV	Wedge $\delta$	Background $\delta$	Isotope
517	$(-4.63 \pm 1.75) \times 10^{-10}$		$^{36}\text{Cl}$
786	$(+1.83 \pm 1.57) \times 10^{-10}$		$^{36}\text{Cl}$
1165	$(+1.48 \pm 0.13) \times 10^{-9}$	$(+0.32 \pm 0.01) \times 10^{-9}$	$^{36}\text{Cl}$
1951	$(+1.11 \pm 0.30) \times 10^{-9}$		$^{36}\text{Cl}$
182	$(-1.11 \pm 0.07) \times 10^{-8}$	$(-0.67 \pm 0.01) \times 10^{-9}$	$^{158}\text{Gd}$

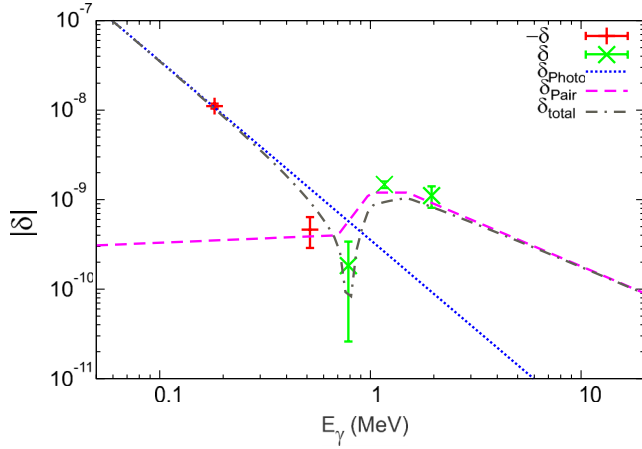


FIG. 2 (color online). Newly measured index of refraction  $|\delta|$  for  $\gamma$  energies up to 2 MeV. The blue dashed curve shows the negative  $\delta$  from the virtual photo effect, which is confirmed by the measured values for lower  $\gamma$  energies. For the positive  $\delta$  of virtual pair creation we used a shape from our dispersion relation calculations. Also the superposition of the two  $\delta$  contributions is shown.

Mössbauer limit  $\Theta_{\text{elas}}$  for the elastic Delbrück scattering  $\sigma_{\text{Delb,elas}}$  and beyond  $\Theta_{\text{elas}}$  for the inelastic Delbrück scattering  $\sigma_{\text{Delb,inels}}$ . Both are shown in Fig. 3.

We now use the dispersion relations [1,8,10] to calculate the real parts  $\delta$  of the refraction index on the left side of Fig. 3. We introduce the outgoing complex forward-scattering amplitude  $A_f(E_\gamma) = A_{rf}(E_\gamma) + iA_{if}(E_\gamma)$ , with the real part  $A_{rf}(E_\gamma)$  and the imaginary part  $A_{if}(E_\gamma)$ . The virtual processes with coherent cross sections  $\sigma_{\text{sca}}$  contribute to the real part  $A_{rf}$ , the real processes with cross sections  $\sigma_{\text{abs}}$ , but also inelastic virtual processes contribute

to the imaginary part  $A_{if}$  of the coherent scattering amplitude

$$A_{rf}(E_\gamma) = \frac{E_\gamma^2}{\pi(4\pi^2\hbar c)^2} \lim_{\epsilon \rightarrow 0^+} \int_0^\infty \frac{A_{if}(E)dE}{E(E^2 - (E_\gamma + i\epsilon)^2)}. \quad (2)$$

By the optical theorem [8,9], the imaginary part of the forward-scattering amplitude is related to the total absorption cross section,  $\sigma_{\text{abs}}(E_\gamma) = 2\lambda A_{if}(E_\gamma)$ , or,  $A_{if}(E) = (\frac{E}{4\pi\hbar c})\sigma_{\text{abs}}(E)$ . We thus obtain

$$A_{rf}(E_\gamma) = \frac{E_\gamma^2}{2\pi^2\hbar c} \lim_{\epsilon \rightarrow 0^+} \int_0^\infty \frac{\sigma_{\text{abs}}(E)dE}{E^2 - (E_\gamma + i\epsilon)^2}. \quad (3)$$

This relation connects the total absorption cross section  $\sigma_{\text{abs}}(E)$  with the forward coherent scattering cross section

$$\frac{d\sigma_{\text{sca}}(\text{forward})}{d\Omega} = |A_{rf}(E_\gamma)|^2. \quad (4)$$

$A_{rf}(E_\gamma)$  is related to the real part of the index of refraction  $\delta(E_\gamma)$

$$\delta(E_\gamma) = \frac{\lambda^2}{2\pi} N_c A_{rf}(E_\gamma), \quad (5)$$

where  $N_c$  is the number of nuclear scattering centers per volume. With these formulas we have calculated the real parts  $\delta_{\text{photo}}$ ,  $\delta_{\text{Compton}}$ , and  $\delta_{\text{pair}}$  for the corresponding three processes photo effect, Compton effect, and pair creation.

Figure 3 shows these three components together with the experimentally measured  $\delta_{\text{photo}}$  from Eq. (1). The strongly rising pair creation cross section and inelastic Delbrück scattering results in a positive  $\delta_{\text{pair}}$ , while the decreasing cross sections of the photo effect and Compton scattering result in a negative  $\delta_{\text{photo}}$  and a negative  $\delta_{\text{Compton}}$ . The

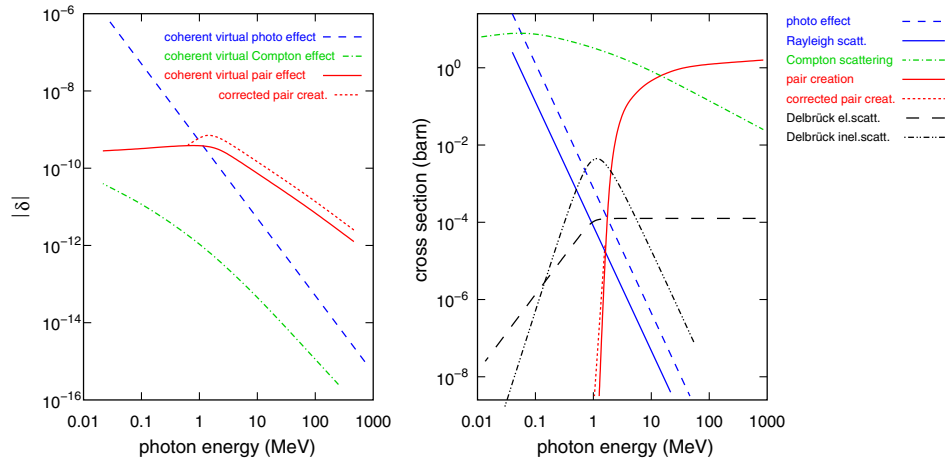


FIG. 3 (color online). Theoretical absolute real part of the index of refraction  $\delta$  (left) and the absorption cross sections (right) in silicon as a function of  $E_\gamma$  from [11]. The three contributions to  $\delta$  from the virtual photo effect, virtual Compton effect, and the virtual pair creation were deduced via the Kramers-Kronig dispersion relations from the corresponding absorption cross sections. The dotted red line (right) shows a recently measured pair creation cross section close to the threshold [14]. Its enhancement over the Bethe-Heitler prediction can again be explained by dispersion relations. We also show the cross section for Rayleigh, elastic Delbrück (virtual pair creation, recoilless), and inelastic Delbrück (virtual pair creation, momentum transfer to the nucleus) scattering. The important new finding is the dominance of the positive  $\delta$  due to virtual pair creation above about 1 MeV  $\gamma$  rays.

surprising result is that  $|\delta_{\text{pair}}|$  becomes larger than  $|\delta_{\text{photo}}|$ , which we wanted to verify experimentally, including the sign change of  $\delta$ .

Historically, J. S. Toll [10] predicted wrongly in 1952 for lead ( $Z = 82$ ) and 1 MeV  $\gamma$  quanta that  $|\delta_{\text{pair}}|$  should be about a factor of  $10^3$  smaller than  $|\delta_{\text{photo}}|$ , discouraging experimentalists from searching for a  $\delta_{\text{pair}}$  contribution. Via dispersion relations they calculated  $\delta_{\text{pair}}$  in first order from the absorptive Bethe-Heitler cross section. When we included also the inelastic, higher-order Delbrück scattering, we obtained the much larger  $\delta_{\text{pair}}$  (red curve), reaching the up-shift of  $\delta_{\text{pair}}$  for 1 MeV to  $1 \times 10^{-9}$ , the range of the measured values. A further increase of  $\delta_{\text{pair}}$  and  $A_{r,f,\text{pair}}(E_\gamma)$  (dashed red curve) occurs, because experimentally a strong enhancement of pair creation close to threshold over the predicted Bethe-Heitler cross section [14] is observed. Since the contribution to  $\delta$  in Eq. (3) becomes very large if  $E$  is close to  $E_\gamma$ , and if  $\sigma_{\text{abs}}$  is steeply sloped as a function of  $E$ , this strong increase of the absorption cross section close to  $2m_e c^2$  increases the value of  $\delta_{\text{pair}}$  significantly. Thus, the newly measured absolute values of  $\delta_{\text{pair}}$  and  $\delta_{\text{photo}}$  can be explained naturally, using the dispersion relations. The higher-order terms with  $Z^4$  and  $Z^6$  make a much stronger contribution to  $\delta_{\text{pair}}$  compared to the first-order term with  $Z^2$ , showing that this corresponds to non-perturbative high field QED, which is addressable by dispersion relations. The value of  $\delta_{\text{pair}}$  is rather constant at low energies, then rises to a maximum between 1 MeV and 2 MeV, followed by a falloff with  $E_\gamma^{-1}$  for higher  $\gamma$  energies. The derivation of Kramers-Kronig dispersion relations is based on a sum decomposition  $\delta_{\text{pair}}(E_\gamma, Z, A, \rho) = \frac{\rho}{A} \times \sum_{n=1}^{\infty} c_n(E_\gamma) Z^{2n}$  assuming a fast convergence of the coefficients  $c_n$ . In case of high fields the convergence assumption is not valid. Although a strict theory does not yet exist, one can expect that the  $Z^4$  term due to the Delbrück cross section in the dispersion integral and higher  $Z^{2n}$  terms become dominant. The present experiment is focused on silicon since technologies for this material for micro-lithographic lens manufacturing are elaborated [15]. However, future experiments will focus on high  $Z$  materials like gold ( $Z = 79$ ). A naive scaling with  $(79/14)^6$  results in a  $\delta_{\text{pair}}$  of  $3 \times 10^{-5}$  in the 1 MeV range. At present the development of small biconvex 1 mm diameter gold  $\gamma$  lenses is underway. We expect a focal length of 3 m and want to determine  $\delta_{\text{pair}}$  accurately by measuring the focal length. As  $\delta_{\text{pair}}$  increases much stronger with  $Z$  compared to  $\delta_{\text{photo}}$ , the switchover energy from positive to negative  $\delta$  is shifted to much lower energies. As shown in Fig. 2, we have a destructive superposition of both contributions of  $\delta_{\text{photo}}$  and  $\delta_{\text{pair}}$  at the switchover energy of about 700 keV for silicon, and  $\delta$  is positive for higher  $\gamma$  energies. For positive  $\delta$ , convex lenses are required for focusing.

We will develop a whole new  $\gamma$  optics tool box:  $\gamma$  lenses for focusing, gold prisms for deflection, and short  $\gamma$  wave guides from gold with total internal reflection. With a

combination of refractive and diffractive  $\gamma$  optics, we can realize very efficient  $\gamma$  monochromatization down to a bandwidth of  $10^{-6}$ , allowing us to address individual nuclear levels up to the neutron binding energy [16]. With the upcoming,  $10^7$  times more brilliant and intense  $\gamma$  beams of MEGa-Ray (Livermore, USA, 2013) [17] and ELI-NP (Bucharest, Romania, 2015) [18], compared to the present worldwide best  $\gamma$  facility HI $\gamma$ S (Duke University, USA), a broad field of new applications with nuclear resonance excitation (radioactive waste management, imaging of  $^7\text{Li}$  in batteries for green energy, production of about 50 new medical radioisotopes with high specific activity for diagnostics and therapy [19], etc.) opens up in nuclear photonics [16]. Also high-resolution nuclear spectroscopy with a few eV resolution [20] and efficient  $\gamma$  astronomy will flourish.

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