# Absolute determination of cross sections for resonant Raman scattering on silicon

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We studied the resonant Raman scattering of x rays in the vicinity of the K absorption edge of silicon. The investigation was carried out at the plane grating monochromator beamline for undulator radiation of the PTB laboratory at BESSY II in Berlin. Cross sections were determined absolutely for a wide energy range of incident photons with small relative uncertainties employing calibrated instrumentation avoiding any reference samples. The experimentally determined values differ clearly from the theoretical ones found in the literature.

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# I. INTRODUCTION

The phenomenon of resonant inelastic x-ray scattering has been studied experimentally more frequently in the past two decades due to the increasing availability of synchrotron radiation allowing for both high flux and highly monochromatic excitation. Inelastic x-ray scattering shows a resonant behavior when the energy of the incoming photons becomes close to the energy of a real excited state of the atom. It evolves gradually into fluorescence until the ionization threshold is surpassed and the fluorescence line is fully developed. The different terminology used to name the observed phenomena reflects the diversity of experimental conditions. Even though the term "resonant inelastic x-ray scattering" (RIXS) would be more appropriate to most of the studies, we prefer the term "resonant Raman scattering" (RRS) because of its widespread use. According to Simionovici et al. [1] and Åberg and Tulkki [2], resonant Raman scattering denotes only one part of resonant inelastic scattering of x rays, where the final state of the scattered photon is discrete. The other part has a Compton-like feature, where the scattered photon and the ejected electron share the available energy in a continuous way. In most of the experiments performed so far the two processes were not separated and can be included under RIXS.

In addition to the interest of comparing absolute experimental results to modern theory, experimental scattering cross sections are of practical importance. For instance, quantitative x-ray fluorescence (XRF) analysis could gain a higher accuracy if the scattering cross sections associated with the spectral background contribution such as RRS originating from relevant matrix elements were better known or available at all. Another important example is the determination of ultra-trace-impurities on a flat substrate such as a Si wafer by means of total-reflection XRF (TXRF). If the energies of fluorescence lines of the impurities are below the absorption edge of a main substrate element and the exciting photon energy is selected to be just below this absorption edge (to avoid the excitation of substrate fluorescence otherwise dominating a TXRF spectrum), an additional strong background is caused by the RRS. Due to the energetic overlapping of the RRS distribution and the fluorescence lines of the impurities, both the detection limits and reliable quantification of the impurities are strongly affected. Up to now, most of the experiments concentrated on the investigation of the RRS on transition metals and rare-earth elements. Some of them included the determination of cross sections [3–7]. Complementarily, the determination of RRS cross sections has still to be performed for many lighter elements.

We studied resonant Raman scattering on silicon. The study is twofold motivated: by the application of TXRF to ultra-trace-element analysis on silicon wafer surfaces and by the need for reliable, preferably reference-free XRF analysis of partially light matrices. Baur *et al.* [8] reported the limitation of the detection sensitivity for aluminum trace contamination on Si wafer surfaces due to resonant Raman scattering. This is the case for an excitation energy just below the Si *K* absorption edge in order to suppress the strong Si *K* fluorescence line. The background under the Al *K* line is mainly caused by RRS on Si. A detailed knowledge of the background, i.e., a modeling with the help of cross sections, would enhance the accuracy of the ultra-trace-analysis.

#### **II. THEORY**

Resonant Raman scattering occurs when the energy of the incident x-ray radiation is smaller than the binding energy of an inner shell of the target atom. An excited state is created which decays via an intermediate state, with an electron in the continuum, into a final state. The final state is characterized by a hole in a higher shell, an electron in the continuum, and the scattered photon. The ejected electron and the scattered photon share the available energy in a continuous way. The process shows a resonant behavior when the energy of the exciting radiation is close to that of a real, excited state of an atom.

A review of the theoretical description of this process can be found in [2,4,9]. Recent theoretical descriptions of resonant Raman scattering deal with radiationless processes [10-12]. In the following we outline the main aspects of the background theory only. The main emphasis is laid upon the

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derivation of an equation that describes the spectral distribution of the RRS, and thus can be used for the fitting procedure in the spectrum evaluation.

Scattering processes are described by the interaction Hamiltonian between the atom and the electromagnetic field of the incident photon. The linear  $\vec{p} \cdot \vec{A}$  term, where  $\vec{p}$  stands for the electronic momentum operator and  $\vec{A}$  for the vector potential of the field, taken in second order, accounts for the resonant Raman scattering. The Kramers-Heisenberg formula [9] provides the double differential scattering cross section for the interaction Hamiltonian. For the case of RRS at the *K* edge with a hole in the *L* shell in the final state, the double differential cross section per unit scattered energy and per unit solid angle can be expressed as follows [4,9]:

$$\left(\frac{d^2\sigma}{d\Omega \, dE}\right) = \frac{r_0^2}{2} \frac{E}{E_0} \frac{(U_K + T_e)(U_K - U_L)}{(U_K - U_L - E)^2 + (\Gamma_K/2)^2} g_{1s,2p} \left(\frac{dg_K}{dT_e}\right)_{U_K + T_e}.$$
 (1)

 $E_0$  is the energy of the incident photon and E that of the scattered photon. They are related due to energy conservation as  $E_0-U_L=E+T_e$  with  $U_L$  for the average binding energy of the L shell and  $T_e$  for the kinetic energy of the ejected electron.  $r_0^2$  is the square of the classical electron radius and  $\Gamma_K$  is the total K-shell width. The oscillator strength  $g_{1s,2p}$  between the  $(1s)^{-1}$  and  $(2p)^{-1}$  hole states and the oscillator density  $dg_K/dT_e$  can be described as [4]

$$\left(\frac{dg_K}{dT_e}\right)_{U_K+T_e} = \frac{\tau_K(U_K+T_e)}{2\pi^2\hbar cr_0}, \quad g_{1s,2p} = \frac{1}{2}\frac{\Gamma_{K\alpha}\hbar c}{r_0(U_K-U_L)^2},$$
(2)

where  $\Gamma_{K\alpha}$  is the width of the  $K\alpha$  fluorescence line and  $\tau_K(U_K+T_e)$  the photoelectric cross section of the *K* shell at the energy  $U_K+T_e$ . The oscillator density  $dg_K/dT_e$  is proportional to the density of empty states in the continuum. If final-state effects are ignored a constant oscillator density can be assumed and an average kinetic energy  $\overline{T_e}$  for the ejected electron may be used. This assumption is justified for measurements with a moderate energy resolution only. For high-energy-resolution experiments the fluctuations in the photoelectric cross section, which are seen in the near-edge and extended-x-ray-absorption fine structure, have to be taken into account. From Eqs. (1) and (2), and assuming a constant oscillator density, the double differential cross section can be written as

$$\left(\frac{d^2\sigma}{d\Omega \ dE}\right) = \Gamma_{K\alpha} \frac{\tau_K (U_K + \overline{T_e})}{8 \ \pi^2 (U_K - U_L)} \frac{E}{E_0} \frac{U_K - U_L + E_0 - E}{(U_K - U_L - E)^2 + (\Gamma_K/2)^2}.$$
(3)

Equation (3) implies that the spectral profile of the RRS emission spectrum is a Lorentzian distribution centered at  $U_K - U_L$ , which is the energy of the  $K\alpha$  fluorescence line. However, the energy conservation between the initial and final states imposes a cutoff on the distribution at the energy  $E_c = E_0 - U_L$ . Due to this cutoff only the left wing of the



FIG. 1. Schematic view of the experimental setup.

Lorentzian distribution remains. Employing both Eq. (3) and the cutoff energy, the spectral distribution of the RRS was calculated and used for the fitting in the spectrum evaluation.

The single differential cross section per unit solid angle is obtained by integrating the energy of the scattered photons between zero and  $E_0 - U_L$  assuming  $U_K - E_0 \gg \Gamma_K/2$ , see [4]. It follows that there is proportionality between the single differential cross section and the difference between the binding energy of the *K* shell and the incident photon energy:

$$\frac{d\sigma(E_0)}{d\Omega} \sim (U_K - E_0)^{-1}.$$
(4)

#### **III. EXPERIMENT**

The present work was performed at the plane grating monochromator (PGM) beamline for undulator radiation [13,14] in the PTB laboratory at the electron storage ring BESSY II. Monochromatized undulator radiation covering the energy range from 78 up to 1860 eV is available here. The beamline operation mode ensures both a high flux and a good suppression of higher undulator harmonics, providing radiation of high spectral purity. In addition, a 1.5- $\mu$ m-thick silicon filter was used 1 m upstream from the PGM focus to suppress stray light contributions far below and just above the silicon *K* edge. The resolving power during the measurements was about 1000. In these experiments resonant Raman scattered radiation was measured under a scattering angle of 90° with respect to the direction of the horizontally polarized incident radiation.

In order to obtain absolute cross sections it is necessary to define and determine all the variables and parameters of the experiment absolutely. The experimental arrangement (see Fig. 1) provides a well-defined beam geometry. Thin samples are positioned in the center of the vacuum chamber. The incoming beam hits the sample of interest at an incident angle of  $45^{\circ}$ . The scattered radiation and the fluorescence radiation are registered at an observation angle of  $45^{\circ}$  by means of a calibrated energy-dispersive Si(Li) detector hav-



FIG. 2. Transmission measurements for the absorption correction involving incident  $(I_0)$  and transmitted  $(I_{tr})$  beam intensities.

ing an active area of 20 mm<sup>2</sup> placed just behind a calibrated aperture. This aperture, having a diameter of  $1.501\pm0.004$  mm, is located at a distance of  $59.4\pm0.2$  mm from the sample center, thus well defining the solid angle of detection.

The angle of 90° between the incident beam and the observation direction of the Si(Li) detector ensures a minimal contribution of Compton and Rayleigh scattered photons in the recorded spectra due to the horizontal polarization of the incident undulator radiation. Apertures 1 and 2 (see Fig. 1) ensure the proper angular alignment of the irradiation chamber with respect to the incident beam and thus that the incident radiant power at the sample position is the same as at a calibrated photodiode behind the chamber. The incident radiant power is determined absolutely by this calibrated photodiode. The spectral response of the diode was measured absolutely as a function of the photon energy during a dedicated calibration in the PTB laboratory at BESSY II [15,16]. To determine the incident radiant power during the recording of the spectra, a reference measurement was performed before and after each recording of a spectrum. During the reference measurements the respective sample was moved out of the incident beam.

In addition, the transmittance of each sample was measured. The energy range for these transmission measurements was chosen to cover the photon energies of interest for the resonant Raman scattered radiation and the photon energies of the respective incident radiation. This range begins at approximately 1000 eV and ends at about 20 eV above the *K* absorption edge of silicon (1839 eV).

# **IV. DATA EVALUATION**

The double differential cross sections  $d^2\sigma/d\Omega dE$  of the resonant Raman scattering can be determined from the number of resonant Raman scattered photons  $dN_{scat}(E)$  with a photon energy between E and E+dE [17]:



FIG. 3. Mass absorption cross sections of silicon.

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$$\frac{dN_{scat}(E)}{dE} = N_{E0} \frac{d\Omega}{4\pi} \varepsilon_{det}(E) \frac{L}{A} \rho \int_{0}^{T} e^{-\gamma \mu(E_{0})x} \\ \times \frac{d^{2} \sigma_{RRS}(E_{0}, E)}{d\Omega \ dE} \bigg|_{\Theta=90^{\circ} \ lin.pol.} e^{-\gamma \mu(E)x} dx.$$
(5)

The total number  $N_{E0}$  of incident photons is determined by the mean incident radiant power, the integration time of the spectra, and the photon energy of the incident radiation. The solid angle of detection  $d\Omega$  is well known as described above. The efficiency  $\varepsilon_{det}(E)$  of the Si(Li) detector as a function of the photon energy E of the scattered photons is known from a dedicated calibration of the detector [18]. Avogadro's number L divided by the atomic weight A of silicon (28.0855) and multiplied by the density  $\rho$  (2.33 g/cm<sup>3</sup>) of silicon converts the cross section into a linear cross section. The density  $\rho$  of the samples is assumed to be the same as the density of the bulk material, as the samples were at least 0.5  $\mu$ m thick. Due to the thickness, a correction taking into account both the absorption of the incident radiation and the self-absorption of the scattered radiation in the sample has to be performed. The attenuation of the incident and outgoing scattered radiation is described by the exponential functions in Eq. (5). They depend on the geometry factor  $\gamma = 1/\cos 45^\circ$ , on the absorption cross sections  $\mu$  of the incident photon energy  $E_0$  and of the photon energy E of the outgoing photon, and on the path length x for the radiation within the sample. The integration of the exponential functions in Eq. (5) along the path x of the radiation in the sample results in an absorption correction factor M[17] as given in the following equation:

$$M(E_0, E) = \int_0^t e^{-\gamma_1 \mu(E_0)x} e^{-\gamma_2 \mu(E)x} dx$$
  
=  $\frac{t}{\gamma \mu(E_0)t + \gamma \mu(E)t} (1 - e^{-\gamma \mu(E_0)t} e^{-\gamma \mu(E)t}).$  (6)

In the soft-x-ray region, values in the literature of absorption cross sections often have unknown uncertainties. Estimates for the uncertainties in the soft-x-ray region range



FIG. 4. (a) Transmittance, (b) thickness, and (c) absorption correction factor M of a nominally 5- $\mu$ m-thick silicon sample.

from 10% to 30% [19,20]. Therefore the literature data for the soft-x-ray region are to be avoided, if possible. To get rid of tabulated absorption cross sections the transmittance of each sample was measured for all photon energies of interest (see Fig. 2) as already mentioned above. However, the use of tabulated absorption cross sections cannot be completely avoided for the determination of the sample thickness. But the relative uncertainties of the absorption cross sections are considerably smaller in the harder-x-ray region and were estimated to be about 2% only. Scholze and Krumrey [21] determined the absorption cross sections of silicon with small relative uncertainties by means of transmission measurements from 100 eV up to 10 keV (see Fig. 3) and theoretical absorption cross sections [22] for energies above 3 keV. The thicknesses t of the samples used in this work were determined by means of these absorption cross sections determined by Scholze and Krumrey [21]. Therewith and with the measured transmittance of the samples the absorption correction factors M can be calculated. Figure 4(a)shows a transmission measurement of a nominally 5- $\mu$ m-thick Si sample. The corresponding thickness of the sample as a function of the photon energy is depicted in Fig. 4(b). The dotted line represents the mean value of the thickness for all photon energies below the silicon K absorption edge. The resulting absorption correction factors for this sample are shown in Fig. 4(c). The double differential cross sections are determined by the following equation, which is obtained from Eqs. (5) and (6):

$$\frac{d^2 \sigma_{RRS}(E_0, E)}{d\Omega \, dE} \bigg|_{\Theta = 90^\circ, \ lin.pol.} = \frac{4 \pi A}{d\Omega \varepsilon_{det}(E) L\rho M(E_0, E) N_{F0}} \frac{dN_{scat}(E)}{dE}.$$
(7)

### **V. SPECTRUM EVALUATION**

Finally, the last value that has to be determined for the double differential cross section is the number  $dN_{scat}(E)$  of scattered photons in the energy range from E to E+dE as recorded by the Si(Li) detector. This was carried out by means of a deconvolution of the measured spectra [Fig. 5(a)]. Since a direct deconvolution cannot be performed, a  $\chi^2$  fitting procedure was employed to determine the raw count rates of the RRS counts detected. Therefore, a theoretical raw spectrum was calculated containing a theoretical RRS distribution [Fig. 5(b)], fluorescence lines, and other spectral features. It is reasonable to calculate this theoretical raw spectrum stepwise. In the present work the channel width of the Si(Li) detector was  $\Delta E=10$  eV.

For fitting a theoretical distribution to the measured spectra, the detector response function has to be taken into account. Therefore we used a theoretical model [18] for the response function and a set of parameters determined experimentally as follows. In dedicated calibration experiments the detector was irradiated directly with monochromatized syn-



FIG. 5. (a) Fit to a measured spectrum of a  $12-\mu$ m-thick Si sample excited by radiation of 1742 eV (gray line, measured spectra; dotted, RRS; black, sum of the fit; dashed, fluorescence lines; dash-dotted, background). (b) Theoretical spectral distribution of the RRS on silicon for incident radiation of 1742 eV.

chrotron radiation of very low flux [23] accessible by special operation modes of BESSY II involving drastically reduced stored electron beam currents. Some Compton or Rayleigh scattered photons were detected by the Si(Li) detector, notwithstanding the exact 90° geometry and the polarization of the incident beam. The energy for these scattered photons is nearly the same as the energy of the incident photons. Therefore, a single energy line at the incident photon energy was added to the composition of the theoretical spectrum. A second feature of the measured spectra is a background caused by bremsstrahlung of photoelectrons produced in the sample. Therefore a spectral distribution for bremsstrahlung given by [24] was added to the theoretical spectrum. To calculate the spectral distribution of the RRS on the silicon L electrons [Fig. 5(b)] the following Eq. (8) was used [see Eq. (3)]. The spectral distribution of the RRS on the silicon M electrons was not added to the theoretical spectrum, since the maximum scattered photon energy is just 2 eV less than the incident photon energy and therewith the main contribution of the KM RRS was taken into account by the single energy line at the incident photon energy already added to the theoretical spectra. The contribution of the low-energetic tailing of the RRS KM distribution in the range of the KL RRS distribution is less than 1% and therefore it was neglected.

$$N_{scat}(E) = H \frac{E}{E_0} \frac{E_{K\alpha} + E_0 - E}{\left(E_{K\alpha} - E\right)^2} \Delta E \quad \text{for } E \le E_{max}.$$
 (8)

The parameters that were fitted are the scaling factor for the height H and the cutoff energy  $E_{max}$ . The cutoff energy should be fixed relative to the energy of the  $L_{2,3}$  absorption edge, but because of the limited energy resolution of the Si(Li) detector and the possibility of a small difference between the energy axis of the beamline and the detector it is necessary to vary this value by a few eV to achieve the optimal agreement with the measured spectra. Integrating the resulting double differential cross sections over the photon energies E of the scattered photons gives the single differential cross section.

$$\frac{d\sigma_{RRS}(E_0)}{d\Omega} = \int_0^{E_{max}} \frac{d^2 \sigma_{RRS}(E_0, E)}{d\Omega \, dE} dE.$$
(9)

The following equation was used to calculate the single differential cross sections by means of Eqs. (8)–(10). The integral was replaced by a summation due to the channels of the Si(Li) detector system.

$$\frac{d\sigma_{RRS}(E_0)}{d\Omega}\bigg|_{\Theta=90^\circ,\ lin.pol.}$$
$$= C \sum_{E_{min}}^{E_{max}} \frac{1}{\varepsilon_{det}(E)M(E_0,E)} \frac{E}{E_0} \frac{E_{K\alpha} + E_0 - E}{(E_{K\alpha} - E)^2} \Delta E.$$
(10)

Some of the determined parameters are summarized in the factor *C*:



FIG. 6. Comparison of the experimentally determined cross sections and the theoretically calculated values; the fitted curve is proportional to the inverse of the energy difference  $(U_K - E_0)$ .

$$C = \frac{4\pi A}{d\Omega L\rho N_{F0}} H.$$
 (11)

The summation in Eq. (10) starts at  $E_{min}$ =1000 eV, since the contribution of the double differential cross section below 1000 eV to the total cross section is less than 0.2%.

## VI. RESULTS AND DISCUSSION

The single differential cross sections were obtained for incident photon energies in a range from 1582 to 1822 eV employing a 12- $\mu$ m-thick Si sample. The values of the single differential cross sections are plotted in Fig. 6. The dependence of the single differential cross sections on the incident photon energy is given by Eq. (4). To prove this dependence the following equation was fitted to the values of the determined cross sections:

$$\left(\frac{d\sigma(E_0)}{d\Omega}\right) = A_0 + \frac{A_1}{(U_K - E_0)}.$$
(12)

It was found that the energy dependence is well described by this equation. The parameters  $A_0, A_1$ , and  $U_K$  were fitted, and therewith a value of 1837±4 eV was determined for the binding energy  $U_K$  of the K shell (1839 eV). By means of Gavrilă's coefficients [25], theoretical values of the double differential cross sections were calculated for three incident photon energies. By fitting Eq. (8) to these values and a summation for all scattered photon energies above 1 keV three values of the single differential cross sections were calculated. Equation (12) was also fitted to these theoretical values (see Fig. 6). The theoretical values are about 20% to 30% below the respective cross sections determined experimentally. This discrepancy is not easy to explain just by the simplifications involved, such as hydrogenic wave functions and the dipole approximation, which Gavrilă and Tugulea [25] have used, but this discrepancy seems to be typical (see [1,26]). This comparison shows that new calculations with current theoretical approaches and numeric modeling are very desirable.

In order to test the evaluation procedure the absorption correction and the thickness determination were carried out for Si samples of various thickness. The cross sections obtained agree very well within their respective uncertainties.

Hall et al. [27] reported a partial cross section for the photon energy of Si  $K\alpha$  (1739 eV) as the incident radiation. This partial cross section is for scattered photons with photon energies in the range from 1634 to 1640.5 eV. By means of Eq. (10) the ratio between this partial energy range and the energy range used in the current work was calculated. Therewith Hall *et al.*'s [27] average partial cross section gives an estimated single differential cross section of  $(4.9 \pm 1.7) r_0^2$  sr<sup>-1</sup>. The fitted equation for the single differential cross sections of the 12  $\mu$ m sample gives a value of  $(5.11\pm0.4)r_0^2$  sr<sup>-1</sup> for the incident photon energy of Si K\alpha. These two values agree well within their respective uncertainties. The uncertainty of Hall et al.'s cross section is quite large; because the RRS feature in their spectra was very small and superimposed by many other effects, they had to perform a complicated procedure to isolate the resonant Raman scattered photons in the spectra.

Another value of the total RRS cross section for Si  $K\alpha$  radiation on Si was reported by Karydas *et al.* [28]. Their value is  $(5.3\pm0.7)r_0^2$  sr<sup>-1</sup> and agrees well also with the cross sections mentioned above.

A total relative uncertainty of 7% was determined for the presented cross sections. This uncertainty is caused by several contributions in Eq. (10). The main contribution is the relative uncertainty of 5% for the factor *H* determined by the fitting procedure. The efficiency  $\varepsilon_{det}(E)$  of the Si(Li) detector has a relative uncertainty of 1.5%, and the solid angle of detection  $\Delta\Omega$  was calculated to have a relative uncertainty of 0.7%.

The determined thicknesses of the samples have an uncertainty of 3% caused by the transmission measurements and the uncertainty of the tabulated absorption coefficients.

The relative uncertainty of the number of incident photons  $N_{E0}$  of 1.5% was caused by the uncertainty of the reference

measurement (0.5%) and the uncertainty of the calibration of the photodiode of 1.5%.

# VII. CONCLUSIONS

The single differential cross sections of resonant Raman scattering on silicon were determined absolutely for a wide energy region of incident photons. Relative uncertainties of about 7% have been achieved. The results of the present work agree well with the few other experimentally values determined by other authors [27,28].

The experimentally determined values clearly differ from the theoretically calculated ones of Gavrilă and Tugulea [25]. These calculations mostly rely on hydrogenic wave functions and the dipole approximation. Hence, new calculations with current theoretical approaches and numeric modeling are desirable.

The method presented in this work to obtain absolute values does not employ any reference samples for the calibration of the experimental setup. Hence, the determination of the cross sections is independent of other fundamental parameters, like fluorescence yields, and their corresponding relative uncertainties. The cross sections absolutely determined will be useful for reference-free x-ray-fluorescence analysis involving silicon containing matrices and substrates. This work will be extended to other elements in order to increase the number of well-known fundamental parameters for reference-free analysis. In addition, a high-resolution spectrometer, already in the progress of commissioning, will be used to study the spectral distribution of the resonant Raman scattering in more detail.

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