




Measurements of electron bremsstrahlung double-differential cross sections for solid targets down to low photon energies: No polarization contribution

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We investigate the question of whether there are any appreciable polarization contributions to the total bremsstrahlung in electron-impact solid thin-film experiments. Our results definitively show no evidence of a polarization contribution. The bremsstrahlung double-differential cross sections at a 90° photon emission angle for collisions of (10–25)-keV electrons with four medium- and high- Z thin-film solid targets are measured, in which the photon energy range is extended down to approximately 3 keV for the solid thin-film experiments. Comparisons of the experimental values with two theories, i.e., ordinary bremsstrahlung (OB) and stripping approximation including polarization bremsstrahlung, are made and good agreement with the OB theory is found. The suppression mechanism of polarization bremsstrahlung in the solid-target experiments will need further theoretical studies.

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Bremsstrahlung is one of the most important physical processes when electrons interact with matter, and reliable bremsstrahlung cross sections are necessary for many fields. However, there is a lack of high-precision experimental cross sections with a wide photon energy range, especially at lower photon energies, for bremsstrahlung by electron impact, which is critical for identifying the contribution of polarization bremsstrahlung [1–4].

According to the classical theory, when an incident electron impacts a target atom, the velocity of the incident electron changes under the action of the Coulomb force of the atom. This process is accompanied by the production of continuous x rays, known as bremsstrahlung. In [5,6] a new mechanism was proposed, which referred to the x rays emitted by the polarized target atom. To differentiate between them, the former is termed ordinary bremsstrahlung (OB) and the latter polarization bremsstrahlung (PB). The incipient theoretical investigation of bremsstrahlung was carried out on the basis of classical electrodynamics [7]. For OB, Tseng and Pratt [8] established a numerical calculation method based on the relativistic partial-wave method, using the exact Dirac wave functions for the electronic scattering states [9], i.e., OB theory. The resulting energy spectrum [10,11] and shape function tables [12,13] are the most reliable theoretical results to date and are theoretical bases of some Monte Carlo programs for simulating bremsstrahlung emissions, such as PENELOPE [14] and GEANT4 [15]. Recently, the BREMS program was developed by Pořkus [16,17] to implement the method from Tseng and Pratt [8], creating a library of bremsstrahlung data for

all elements over a wide range of incident electron energies and a dense energy grid. In contrast, the theoretical study of PB is still underdeveloped. The stripping approximation (SA) for calculating the total bremsstrahlung including the PB contribution was proposed by Avdonina and Pratt [18] and Korol *et al.* [19].

In order to verify the accuracy of the theory and to explore whether PB exists in the total bremsstrahlung, some researchers have performed experimental measurements for bremsstrahlung double-differential cross sections (DDCSs) and compared them with theoretical results. The definitive contribution of PB for rare-gas atoms was confirmed by comparisons between the measured bremsstrahlung DDCSs by Portillo and Quarles [20] and the OB and SA theories. It was found that the measured data followed the same trend as the SA theory, but with some obvious differences in magnitude, which have not been quantitatively explained yet [20]. The polarization bremsstrahlung was also verified for Ar atoms by Prajapati *et al.* [21].

However, the question of whether there are any appreciable PB contributions to the total bremsstrahlung spectra in experiments involving solid targets has remained inconclusive up to now. Several works [22–26] have suggested that the PB contribution was not found in the interactions of electrons with the solid thick targets, as the experimental spectra were in general agreement with the OB theoretical results. Nevertheless, a different view that PB is obvious in the low-photon-energy region has been put forward [3,27–30]. For instance, the PB contribution for the Pb target is approximately 44% at a photon energy of 1 keV [30]. We have measured the bremsstrahlung spectra for 11 thick targets with (5–25)-keV electron impact and also found good agreement between the measurements and the simulated results by the

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PENELOPE code containing only the OB theoretical values [31,32].

For identifying whether PB exists in the total bremsstrahlung for electrons colliding with solid targets, the best choice is to measure the bremsstrahlung DDCSs, especially at lower photon energies, and directly compare them with the theoretical DDCS values. It is obvious that the preparation of thick targets and the associated experiments are relatively easy. However, the experimental bremsstrahlung spectra for thick targets are unlikely to be directly compared with the theoretical bremsstrahlung DDCSs, due to the complicated physical processes of electrons and photons that occur in thick-target experiments. Ideally, interactions between electrons with sufficiently thin-film targets are less affected by multiple scattering and energy loss, etc., and the measured bremsstrahlung DDCSs can be directly compared with the theoretical results. However, due to the difficulties in the thin-film experiments, for example, the thin-film target preparation and the accurate determination of target thickness, absolute measurements of bremsstrahlung DDCSs are still lacking.

The earliest work on absolute measurements of bremsstrahlung DDCSs can be found in [33]. The experimental cross sections for Al and Au with 50-keV electron impact have been measured using a scintillation spectrometer by Motz and Placious [33], with uncertainties of 10%–15%, and compared with the theoretical values of Sommerfeld [34] and Kirkpatrick and Wiedmann [35]. General agreement between the measured data with uncertainties of about 5%–20% and theoretical calculations based on the program of Brysk *et al.* [36] has been observed by Rester *et al.* [37] for Al and Au targets with collisions of 50- and 200-keV electrons. In the work of Quarles and Heroy [38] and Ambrose *et al.* [39], the experimental bremsstrahlung DDCSs for (50–140)-keV electron bombardment of several thin-film targets were in agreement with the OB theory of Pratt and co-workers [12,40,41] within an experimental uncertainty of 22%. The experimental uncertainty was mainly derived from the uncertainty of the target thickness [38,39]. Williams and Quarles [4] and García-Alvarez *et al.* [42] have given the latest experimental bremsstrahlung DDCSs for dozens-of-keV electrons colliding with thin-film targets. It is noteworthy that only the measurement data at higher photon energies (greater than 10 keV) have been reported in the aforementioned studies on cross sections of bremsstrahlung, but no data at lower photon energies are available. In fact, the bremsstrahlung DDCSs in the low-photon-energy range deserve extra attention, as it has been pointed out that PB is most likely to occur in this region [1–4], i.e., they are more conducive to determining the PB contribution.

We have also reported absolute measurements of bremsstrahlung DDCSs for two low-*Z* elements C and Al and compared them with OB and SA theoretical results and some measured data of García-Alvarez *et al.* [42,43]. In general, the OB theory was found to be in better agreement with the measured data than the SA theory and the agreement became better as the photon energy increased. However, there were obvious differences between the measured data and the theoretical values of OB and SA, especially in the low-photon-energy region. Therefore, more extensive measurements are

needed to definitively verify whether the PB contribution exists in the electron-solid interaction.

In this work the bremsstrahlung DDCSs for photons radiated at 90° for four thin-film targets (Ti, Cu, Ag, and Au) with (10–25)-keV electron collisions are measured. Ag and Au are the benchmark elements in the OB theory and their theoretical cross sections have better accuracy. The photon energy range is extended down to as low as approximately 3 keV for the solid thin-film experiments. Our accurate measurement provides a definitive answer for the question of whether there are any appreciable PB contributions in the electron-solid interaction.

The thin-film targets used in this work were prepared at the China Institute of Atomic Energy (CIAE) [44]. To cope with the difficulties of making and transporting self-supporting thin-film targets, the four targets were made in a double-layer structure with a thin carbon substrate. A design of the target structure was carried out using the Monte Carlo PENELOPE code, taking into account the energy loss of electrons in the target and the proportion of the bremsstrahlung from carbon substrate to the total bremsstrahlung of the double-layer target. The purity of the Au and C target was 99.99% and that of the other targets was 99.9%. The carbon film, prepared by the ac carbon arc method, was fixed in the central opening of the circular carbon target frame. The carbon target frame was 18 mm in diameter, 0.5 mm thick, and had a central opening of 4 mm in diameter. The four target films were prepared by depositing on the carbon films using the resistance heating vacuum evaporation method. The target thicknesses were determined at CIAE using an ultramicroelectronic balance (XP2U, METTLER TOLEDO, Switzerland) with a readability of 0.1 μg .

Since the accuracy of the bremsstrahlung DDCSs is directly dependent on the accuracy of the target thickness, we have also measured the target thicknesses using Rutherford backscattering spectrometry (RBS). The RBS measurements were carried out using the 3-MV Tandemron accelerator installed at Sichuan University [45] (see the Supplemental Material [46] for details). The RBS spectra were analyzed using the SIMNRA program [47]. The RBS uncertainty was less than 5%, mainly from the statistical uncertainties and the uncertainties of incident charges. The differences between the results of RBS and the weighing method were less than 5.2%. The thicknesses measured by RBS were 342.7 ± 14.7 , 334.7 ± 6.9 , 354.0 ± 7.3 , 254.9 ± 5.2 , and 141.1 ± 2.9 (in units of 10^{15} atoms/cm²) for the C substrate and the Ti, Cu, Ag, and Au thin-film targets, respectively.

The experimental setup has been described in Ref. [43]. A well-focused electron beam provided by a scanning electron microscope (KYKY-2800B) was directed vertically onto the center of a thin-film target placed on a copper holder at an angle of 45° to the horizontal. Compared to the thick-target experiment, the depth of the Faraday cup has been extended to minimize the effect of scattered electrons and x rays from the bottom of the Faraday cup. The diameter of the electron beam spot on the target was approximately 2 mm. The resulting x rays were collected by a horizontally placed silicon drift detector (SDD) (XR-100SDD, Amptek, USA). Due to the minimal absorption of the ultrathin C2 window consisting of 40 nm of Si₃N₄, a 30-nm Al coating, and a 15- μm Si grid,

the use of XR-100SDD was beneficial for the detection of low-energy photons. Considering that the scattered electrons passing through the C2 window would interfere with the normal operation of the detector and also cause additional bremsstrahlung, a pair of permanent magnets with 800 G was positioned in front of the SDD to deflect the scattered electrons.

We have performed the efficiency calibration of the SDD using two methods for different energy regions and have found that the experimental efficiency values agree well with the theoretical efficiency curve [43]. The number of incident electrons was measured by the ORTEC 439 digital current integrator, which was calibrated using a KEITHLEY 6430 SourceMeter, which provided a constant current of various intensities from 0.1 to 1 nA with an accuracy of 0.15%. The results showed that the uncertainty of ORTEC 439 was less than 1%. A bias voltage of -100 V was set on each of the two holes (i.e., the incident hole for electrons and the detection hole for the SDD) of the Faraday cup to prevent the low-energy electrons from escaping. The x-ray energy spectra were recorded with and without the target film at the center of the carbon target frame and it was found that the background accounted for less than 1% of the measured spectrum.

The parallelized PENELOPE (version 2008) code was used for the calculation of the escape rate for backscattered electrons from the two holes of the Faraday cup and for the simulation of the experimental bremsstrahlung for the correction of the measured cross sections. The geometrical configuration in the Monte Carlo simulation is identical to the experimental setup, i.e., including the target structure, target holder, and Faraday cup and using their accurate dimensions. The cross sections for bremsstrahlung and characteristic x rays in the PENELOPE's database are from the OB theory [8,10,13] and the distorted-wave Born approximation theory [48,49], respectively.

The escape rates of backscattered electrons calculated by the PENELOPE were 0.18%–1.42%, 0.32%–1.76%, 0.57%–2.05%, and 0.59%–1.98% for Ti/C, Cu/C, Ag/C, and Au/C targets for the incident electrons at different energies, respectively, which were used for the correction of the incident charges.

A typical experimental x-ray spectrum (including bremsstrahlung and characteristic x rays) of a Cu thin-film target with 20-keV electron impact is given in Fig. 1. The small peak at 1.7 keV may come from interference of silicon material in the SDD as indicated by the manufacturer [50]. The peaks at 3.7 keV are possibly produced by the interactions of scattered rays with the calcium in the targets, the effect of which has been verified to be inappreciable by the Monte Carlo simulations.

In the data processing, the multiple scattering of electrons, backscattering from the carbon substrate, and absorption of photons in the target should be considered. In this work, the correction factor C_f is given by the ratio of σ_{th} and σ_{MC} ,

$$C_f = \frac{\sigma_{\text{th}}}{\sigma_{\text{MC}}}, \quad (1)$$

where σ_{th} and σ_{MC} are the scaled bremsstrahlung DDCSs obtained by interpolation from the PENELOPE OB theoretical database and calculated by simulating real experimental con-

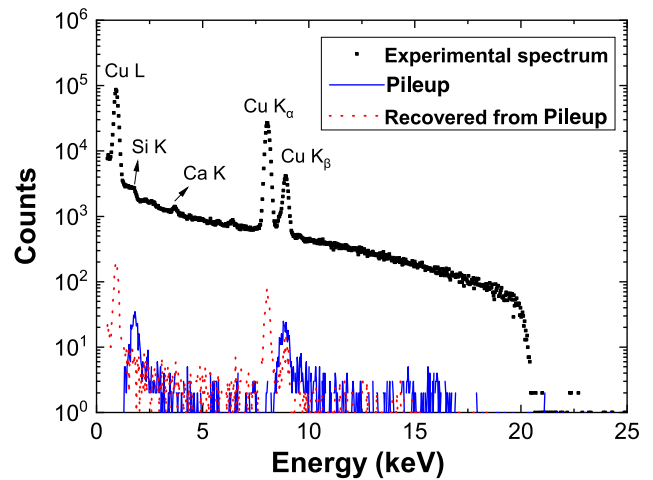


FIG. 1. Experimental spectrum for the Cu/C target with 20-keV electron impact. The pileup spectrum and the recovered spectrum are obtained by the PUC code.

ditions using PENELOPE, respectively (see the Supplemental Material [46]).

The correction factors C_f for the measured bremsstrahlung DDCSs of (10–25)-keV incident electrons for the four thin-film targets and the error bars are given in the Supplemental Material [46]. The uncertainties for the correction factors are between 2.0% and 10.5%, mainly from the statistical uncertainty of the Monte Carlo simulations.

In order to minimize the pileup effect, the experimental count rates were always kept below 500 counts/s in this work. Also, a computational code PUC based on the Monte Carlo algorithm of Ref. [51] and the sampling method by Walker [52] has been developed to correct the pileup effect. The pulse shape of the SDD recorded by an oscilloscope was discretized with a time step of $1 \mu\text{s}$ and the resolution time was determined to be $3.5 \mu\text{s}$. As an example, the pileup spectrum and the recovered spectrum obtained by the PUC for the measured bremsstrahlung spectrum of a 20-keV electron colliding with the Cu/C target are also shown in Fig. 1. All experimental spectra have been corrected by PUC and the pileup effect on the measured results was found to be less than 1%.

The experimental bremsstrahlung DDCSs can be calculated as

$$\sigma_{\text{expt}} = \frac{N_x / \Delta E}{N_e (t / \cos \alpha) \Omega \varepsilon} C_f, \quad (2)$$

where N_x is the measured count of bremsstrahlung photons within an energy range of ΔE , N_e is the count of the incident electrons, t is the number of atoms per unit area, α is the angle between the target surface normal and the incident direction of the electron beam, Ω is the solid angle subtended by the detector, and ε is the intrinsic detection efficiency of the SDD. To facilitate the comparison with theoretical scaled DDCSs, all experimental data were transformed to scaled DDCSs (see the Supplemental Material [46]). For the double-layer targets, the measured bremsstrahlung contains the contributions from the target film and the carbon substrate. To obtain the counts of bremsstrahlung photons N_x generated from the target film, the bremsstrahlung of the self-supporting carbon

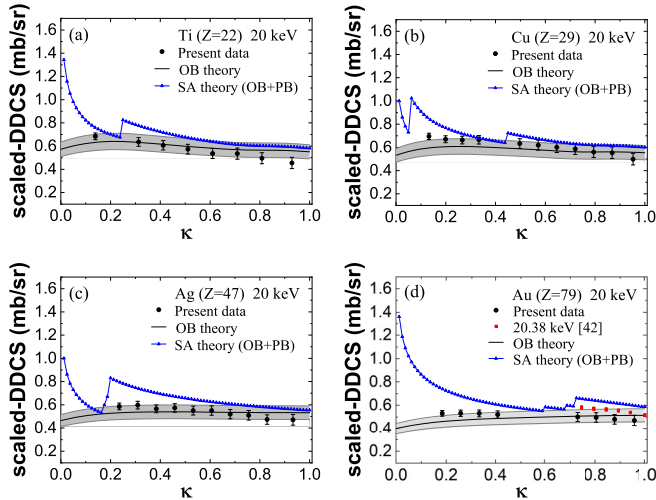


FIG. 2. Scaled DDCSs of bremsstrahlung at 90° photon emission angle for the elements Ti, Cu, Ag, and Au with 20-keV electron impact.

film normalized by the number of incident electrons is subtracted from the total bremsstrahlung counts and the thickness of the self-supporting carbon film is the same as the thickness of the carbon substrate.

The RBS analysis revealed that oxidation occurred in the Ti film, with the number of oxygen atoms accounting for approximately 16% of the total number of atoms. The ratios of theoretical bremsstrahlung cross sections for a titanium oxide molecule [14] and titanium atom were then used to correct for the effect of oxygen atoms in the experiment, and the difference between the cross sections before and after the correction was about 2%.

Figure 2 presents the comparisons of the measured bremsstrahlung DDCSs for the four elements with 20-keV electron impact with the OB and SA theoretical results; the comparisons for 10-, 15-, and 25-keV electron impact are given in the Supplemental Material [46]. The κ is the ratio of the bremsstrahlung photon energy to the incident electron energy. The uncertainties of the measured data represented by error bars in the figure are 3.0%–11.0%, which mainly come from the correction factor (2.0%–10.5%), the statistical counts of the photons (0.3%–2.2%), and the detection efficiency of the SDD (less than 2%). The uncertainties of target thickness (less than 5.2%) and incident charges (less than 1%), as systematic uncertainty, are not included in the error bars. The OB theoretical data were extracted from the PENELOPE database by interpolation and have an uncertainty of 11%

represented by the error bands in the figures, mainly caused by the scaled cross sections (10%) and the shape functions (5%). The calculations for cross sections based on the SA theory that takes into account the PB contribution were performed according to Ref. [4].

For the four elements, in general, the measured cross sections for bremsstrahlung generated by (10–25)-keV electrons are in very good agreement with the OB theoretical results. In contrast, the SA theoretical results are overall higher than the experimental data and there are significant differences in shape, especially in the low-photon-energy region.

For the element Au [Fig. 2(d)], the measured data with 20.38-keV incident electrons of García-Alvarez *et al.* [42] are plotted to compare with our experimental data. It is clear that both sets of measured data are in accordance with the OB theory within the uncertainties and obviously lower than the SA theoretical values. García-Alvarez *et al.* [42] reported the measurements in the higher-photon-energy range ($\kappa > 0.74$), while the differences between the SA and OB theoretical values are more pronounced in the lower-photon-energy region ($\kappa < 0.6$). In view of this, the data in the lower-photon-energy range (down to approximately 3 keV) in this work are critical for judging the two theoretical results as well as for determining whether the PB contribution exists in solid-target experiments.

In conclusion, the double-differential cross sections of bremsstrahlung at 90° photon emission angle in the photon energy range down to approximately 3 keV for four medium and high-Z elements with (10–25)-keV electron impact have been measured. The RBS was used to accurately measure the thicknesses of thin-film targets, circumventing the large uncertainties that will be propagated to the experimental data. Corrections have also been made for the multiple scattering of electrons in the target, backscattering from the carbon substrate, and the signal pileup. The experimental cross sections were found to be in very good agreement with the OB theory, while there were obvious differences in amplitude and shape from the SA predictions that take the PB into account. The results presented in this Letter definitively show no evidence for the PB contribution in solid-target experiments. In comparison with the rare-gas experiment, the suppression mechanism of polarization bremsstrahlung in the solid-target experiment should be further investigated theoretically and quantitatively.

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