Test of Wannier Threshold Laws: Double-Photoionization Cross Section in Helium

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An extensive study of the threshold law for the cross section of double photoionization in helium is presented. It provides quantitative information about the Wannier exponent α, which agrees with the theoretical prediction. In addition, the hitherto unknown constant of proportionality σ_0 for the cross section σ^{++} was determined as well as the energy range of validity, which was found to be smaller than expected.

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The threshold behavior of processes such as double photoionization belongs to the fundamental processes of atomic physics: It requires a solution of the three-body Coulomb problem where the boundary conditions for two continuum electrons have to be included. Since the classical work of Wannier,[1] numerous theoretical studies have been made on near-threshold ionization of atoms by electron of photon impact.[1-13] The various theories yield predictions for three different observable statements: (i) the energy dependence of the cross section, (ii) the energy sharing of the two outgoing electrons, and (iii) the angular correlation of these electrons. In the present Letter we report on the cross section for double photoionization in helium. Wannier theory predicts, in the energy range just above threshold,

$$\sigma^{++}=\sigma_0 E_{\rm exc}^{\alpha}, \qquad (1)$$

with α, the Wannier exponent, being equal to 1.056 for $Z=2$, where Z is the charge of the final ion, $E_{\rm exc}$ the excess energy, and σ_0 the constant of proportionality, i.e., the value of σ^{++} at $E_{\rm exc}=1$ eV.

By an extensive study of this threshold law, we mean an investigation of three problems: (i) the absolute value of σ_0, (ii) the value of the exponent α, and (iii) the energy range of validity for $E_{\rm exc}$ in Eq. (1). Until now the absolute value σ_0 has been determined only for photodetachment from negative ions [experiment for He^- (Bae, Coggiola, and Peterson[14]), K^- (Bae and Peterson[15]); theory for H^- (Ref. 13)]. The experimental determination of the exponent α also concentrated on photodetachment from negative ions [for H^- (Donahue *et al.*[16])] or on single ionization following electron impact.[17,18] Recently, Lablanquie *et al.* studied double photoionization in argon.[19] A superimposed resonance structure, however, prevented the precise check of the value $\alpha=1.056$ as well as the investigation of the other two problems related to the cross section σ^{++}. The energy range of validity for the threshold statements has found considerable interest, theoretically[8,11] as well as experimentally.[16-18,20-23] These investigations show that different energy ranges of validity hold for the three different observable statements of the threshold law given above.[8] Screening effects may cause quite wide validity ranges. For the case of the σ^{++} cross section in helium several electronvolts can be expected.[8,11]

In this Letter we report the first extensive study of the double-photoionization cross section σ^{++} in helium in the threshold region up to 83-eV photon energy. The experiment was performed at the BESSY storage ring on the toroidal grating monochromator TGM4. The band pass was set at approximately 130 meV at 80-eV photon energy, and higher orders from the grating were proved to be negligible for photon energies of interest to the experiment. Ion analysis was carried out with a versatile pulsed-field time-of-flight e/m analyzer developed for working at the conditions of a quasicontinuous photon beam (multibunch mode of the electron storage ring with bunches of 50 ps FWHM separated by 2 ns). The ion analyzer and its performance will be described in a forthcoming publication.[24] The target pressure was 2×10^{-5} mbar. Absolute values for σ^{++} have been established by the measurement of the ratio of He^{++} to He^+ ions and scaling it to the known total absorption cross section.[25] For given fields in the ion analyzer, singly and doubly charged ions have different transmission and detection efficiency. This difference has been taken into account with the information from additional measurements with electric fields adapted to the differently charged ions. The ion analyzer was operated in a cyclic mode with 9-μs repetition time. Different e/m values were accumulated in different gates by the electronics. Each gate accepted at most one ion per cycle. For this mode of operation a formula has been derived on the basis of probability distributions which allows for the exact correction of multiple events per cycle. Such a correction becomes important for counting rates above 2 kHz.

Close to threshold a serious complication arises in the small He^{++} signal which is disturbed by the following: (i) H_2^+ ions which have practically the same e/m value as He^{++} and originate from the small amount of water in the residual gas. (ii) Some H^+ ions (also from water) which still enter the He^{++} time window. This is a particular effect of the quasicontinuous photon beam, which allows for the production of ions during the acceleration period of the ion analyzer. Such ions do not gain the full energy and their detection is therefore delayed. (iii) He^{++} ions caused by stray light (with unknown energies) from the monochromator. The first two disturbances were investigated by the measurement of the H_2^+-to-H^+ ratio without helium in the equipment. This ratio was found to be constant in the energy range of interest. The stray-light contribution to He^{++} was determined for various energies from photoionization processes below the double-ionization threshold. This contribution could be accounted for by an 11% larger H_2^+-to-H^+ ratio. With the assumption of a constant stray-light contribution also above the double-ionization threshold, this enlarged and constant H_2^+-to-H^+ ratio has been applied to correct all He^{++} data. For this purpose, each experimental point is based on two successive ion measurements. First, at the selected photon energy above the double-ionization threshold, ions are counted in three different time windows corresponding to H^+, He^{++} with disturbances, and He^+, respectively. Second, at 78.5-eV photon energy the corresponding disturbance in the He^{++} window was ascertained. In addition to these two successive ion measurements for each experimental point an energy calibration was performed. An accurate energy calibration is necessary for the establishment of the double-photoionization cross section in the threshold region. For this purpose the absorption fine structure near the L_3 edge of aluminum at 72.699 eV [26] was used. This provided an accuracy of approximately 5 meV for the relative energy scale of our data points. The absolute energy scale was checked by means of electron spectrometry with photolines of helium produced by first- and second-order light of the monochromator. This method established an accuracy of approximately 8 meV. Before and after the measurements for the double-photoionization cross section in the threshold region, the He^{++}/He^+ ratio at 100-eV photon energy was determined in order to check the general performance of the apparatus. Its value of 2.27(5)% agrees well with earlier data 2.4(2),[27] 2.8(2),[28] and 2.8(2).[29]

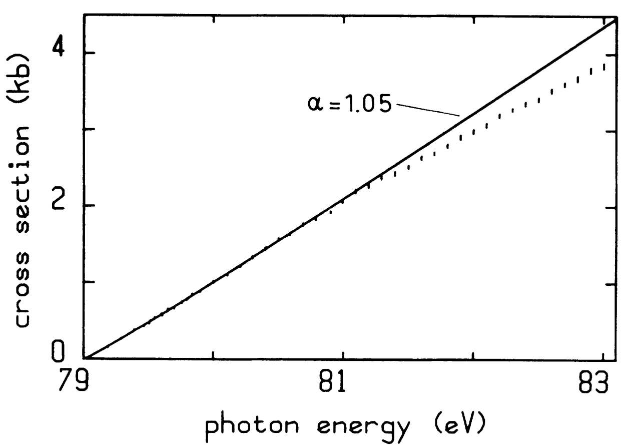

FIG. 1. Double-photoionization cross section in helium from threshold to 83-eV photon energy. The experimental data are given as points with error bars. Disturbing contributions to He^{++} have been subtracted as described in the text. Depending on the actual photon flux the disturbing contributions amount to about 10 counts/s. At 79.7-eV photon energy the He^{++} signal becomes equal to the disturbing contributions. In order to minimize the statistical error, more than 10^4 He^{++} events were accumulated for each point. The solid line represents the least-squares fit of Eq. (1) to the experimental points up to 81-eV photon energy with $\alpha = 1.05$.

The threshold cross-section data are plotted in Fig. 1 and on a shorter but enlarged energy scale in Fig. 2. The error bars contain the statistical error of the counting rate, the uncertainty in the disturbances in the He^{++} signal, and the uncertainty in the relative energy scale. The solid line in both figures represents in a limited energy range a least-squares fit of the experimental data by the power law of Eq. (1). Because of the small difference between a linear behavior ($\alpha = 1$) and the expected $\alpha = 1.056$ threshold law the different steps of the fitting procedure are explained in the following. The most important information comes from the dependence of the free fit parameters on the data points up to a given photon energy. Several distinct fitting procedures shall

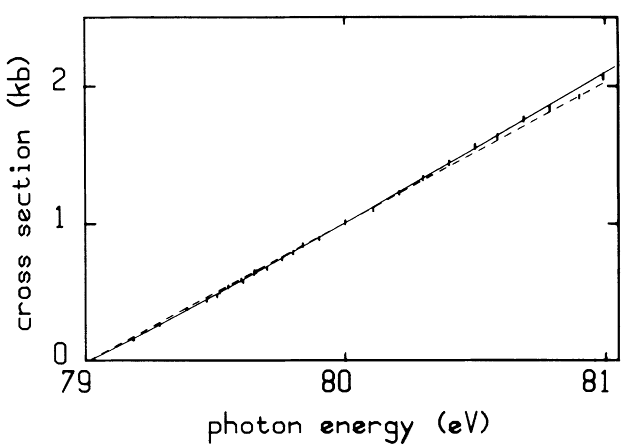

FIG. 2. Double-photoionization cross section from threshold up to 81-eV photon energy (enlarged view from Fig. 1). Solid line: least-squares fit to the experimental points according to Eq. (1), with $\alpha = 1.05$, $\sigma_0 = 1.02 \times 10^{-21}$ cm^2, and $E_{th} = 79.013$ eV; dashed line: straight line with same values for E_{th} and σ_0.

be discussed. The first case concerns a fit of the experimental data by a straight line [$\alpha = 1$ in Eq. (1)] where E_{th} and σ_0 were used as free fitting parameters. Here the fit result for E_{th} already provides a clear criterion about the quality of the linear threshold law because it can be compared with the well-known value from optical data (79.003 eV).[30] The straight-line fit yields over the whole energy range of interest a significantly higher value of E_{th} [for example, $E_{th} = 79.034(5)$ and 79.042(3) eV for ranges of the photon energy up to 80 and 81 eV, respectively]. This discrepancy excludes a threshold law with $\alpha = 1$. In a second fitting procedure Eq. (1) was used with E_{th}, σ_0, and α as free fit parameters. In the first 600 meV above the threshold the experimental uncertainties still yield fluctuations in all three fitting parameters. However, for higher photon energies up to 81 eV all three fitting parameters become rather stable (compare, for example, the behavior of the exponent α, Fig. 3, lower part). Their values are $E_{th} = 79.013(5)$ eV, $\sigma_0 = 1.021(5) \times 10^{-21}$ cm^2, and $\alpha = 1.042(5)$. If we take into account the uncertainty of the monochromator calibration of 8 meV, there is good agreement between this value for E_{th} and that from the optical data (79.003 eV), which confirms our energy calibration and allows us to fix E_{th} in further fitting procedures. From the lower part of Fig. 3 it is also clear that the α values are quite stable around a mean value which is larger than 1 up to a photon energy of about 81 eV. Above this energy a continuous decrease of the α values starts, which is accompanied by a continuous increase in the values of E_{th} and σ_0. This behavior suggests that the threshold law of Eq. (1) is valid only up to about 2 eV above threshold. In order to improve the accuracy of the exponent α, further fitting procedures were performed keeping E_{th} fixed (middle part of Fig. 3) and keeping both E_{th} and the quite insensitive σ_0 value fixed (upper part of Fig. 3). The exponent α becomes stable and definitely larger than 1 in the whole energy range up to approximately 81-eV photon energy. For this threshold region we are able to quote the following final values: $E_{th} = 79.013(10)$ eV, $\sigma_0 = 1.02(4) \times 10^{-21}$ cm^2, and $\alpha = 1.05(2)$. Here the error bars contain additional uncertainties; for E_{th} from the monochromator calibration; for σ_0 from the absolute normalization with the absorption data[25]; for α from the mutual influences through the uncertainties in E_{th} and σ_0.

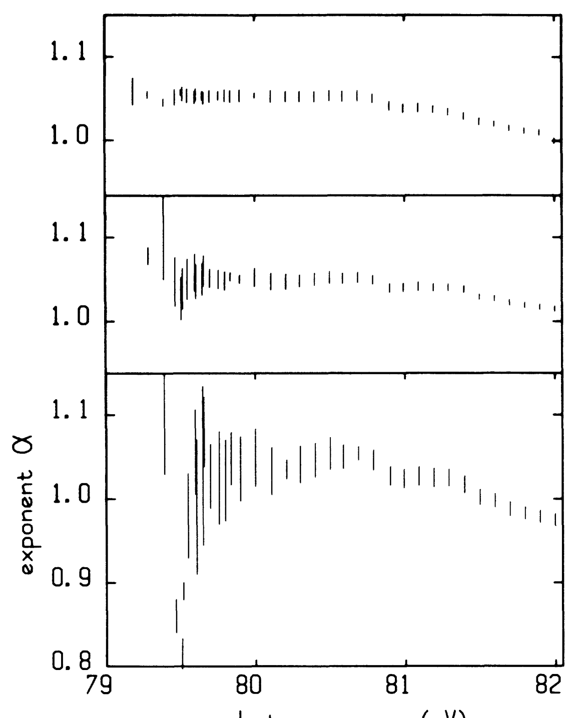

FIG. 3. Wannier exponent α as function of the data points included in the fitting procedure up to a given photon energy. Three different fit results are shown which are all based on Eq. (1). Lower part: E_{th}, σ_0, and α are taken as free parameters; middle part: E_{th} is fixed at 79.013 eV, but σ_0 and α are taken as free parameters; upper part: E_{th} and σ_0 are fixed at 79.013 eV and 1.02×10^{-21} cm^2, respectively, but α is still a free parameter.

It should be noted that the reduced χ^2 value of each fit may also serve as an indicator for the quality of the fit. We therefore determined the reduced χ^2 values of two completely independent fits, one for $\alpha = 1$ and the other for $\alpha = 1.05$, with free parameters E_{th} and σ_0. If we start at threshold, both χ^2 values are similar. However, towards 81-eV photon energy they separate to $\chi^2(\alpha = 1) = 1.38$ and $\chi^2(\alpha = 1.05) = 0.85$, respectively. The latter value supports again the nonlinear threshold law of Eq. (1).

The deviation of the cross section from a linear behavior can be seen also by our comparing in Fig. 2 the experimental data with a nonlinear threshold law (solid line with $\alpha = 1.05$) and with a linear threshold law (dashed line with $\alpha = 1.0$). Equation (1) gives, at 1.0 eV above threshold, for all values of α, the same cross section; therefore, the two lines intersect at $E_{exc} = 1.0$ eV. However, $\alpha > 1.0$ requires for $E_{exc} < 1.0$ eV a cross section smaller than the dashed line ($\alpha = 1.0$), while for $E_{exc} > 1.0$ eV it has to be above that line. The experimental data show this behavior.

Summarizing we are able to conclude the following: Our experimental value $\alpha = 1.05(2)$ confirms the theoretical prediction, $\alpha = 1.056$. For σ_0 there is no direct calculation available. However, calculations of Carter and Kelly[31] and Tiwary,[32] which concentrated on the σ^{++} cross section of helium over a wide energy range above the threshold, provide for an estimate. From the figures in their publications a threshold value σ_0 of approximately 1.1×10^{-21} cm^2 (Ref. 31) or 0.6×10^{-21} cm^2 (Ref. 32) can be extracted. The first value agrees well with our experimental result. It should be noted that this value is considerably smaller than those for double photodetachment ($>10^{-20}$ cm^2).[13-15] Concerning the range of validity of the cross-section threshold law in helium, our experiment yields the result that Eq. (1) is valid up to approximately 2-eV excess energy above threshold. This value is smaller than the several electronvolts which could be expected from theoretical studies[8,11] on the basis of the uniform experimental energy distribution of the two escaping electrons after electron-impact ionization, 3.6 [20] and 5.5 eV.[21]

The present data provide the basis for decisive tests of theoretical treatments for the threshold behavior of the double-photoionization cross section in helium. Especially, this concerns the still outstanding direct calculation of the constant of proportionality σ_0 and a direct theoretical estimate of the energy range of validity for the case of double photoionization.

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