Absolute total-cross-section measurements for intermediate-energy electron scattering on C_2H_2 and CO

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Absolute total-cross-section measurements have been performed on the e^- -C₂H₂ and e^- -CO systems in the energy range 400—2600 eV. An apparatus with linear transmission technique was used. The estimated total experimental errors are about 5%. The results have been compared with available experimental and theoretical results. No previous data for e -C₂H₂ have been found in the literature for impact energies above 400 eV.

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I. INTRODUCTION

Knowledge of the total cross sections for electron scattering (hereafter abbreviated as TCSES) from atoms and molecules is very important in testing various models of the electric and magnetic interactions. The data of TCSES are also important in developing subjects such as astrophysics, plasma physics, atmospheric physics, and chemical physics. The first measurements of TCSES from atoms and molecules date back to the beginning of this century [1,2]. Prior to 1977, Bransden and McDowell reviewed the theoretical [3] and the experimental work [4] of TCSES in the intermediate energy range. Carbon monoxide (CO) is important chemical raw material and fuel gas. Previous TCSES experiments from CO were carried out at electron energies up to 500 eV by Kwan et al. [5], Sueoka and Mori [6], and Kanik, Nickel, and Trajma [7], except those that were listed below 400 eV by Karwasz et al. [8], and above 500 eV by Karwasz et al. [8] and Garcia, Aragon, and Campos [9]. Acetylene (C_2H_2) is an important industrial chemical (found in, e.g., benzene, vinyl chloride, acrylic acid, and esters) [10] and is the essential catalyst in the high-temperature welding for the present industry. By comparing with the total cross sections of hydrocarbon molecules such as acetylene, ethylene, and methane, one can obtain knowledge about the different carbon-to-carbon bonds and the physics of those hydrocarbons (such as C_2H_2 , C_2H_4 , C_2H_6 , CH₄, etc.). In the TCSES of C_2H_2 , previous experiments were carried out at electron energies up to 400 eV by Sueoka and Mori [11]. The lack of TCSES measurements above 400 eV for C_2H_2 and the obvious difference in the TCSES of CO in the higher-energy range have prompted this work.

The N_2 molecule, acetylene, (C_2H_2) molecule, and CO molecule belong to the isoelectronic molecule class (14 electron systems). The similarities in the energy dependence of TCSES for some isoelectronic molecules have been investigated by means of experiments by Zecca, Karwasz, and Brusa [12], Kwan et al. [13], and Karwasz et al. [8] and theoretical calculations by Jain and Baluja [14]. A comparison between the TCSES for CO and the TCSES for C_2H_2 molecules is made in this work.

II. EXPERIMENTAL SETUP

The experimental setup was arranged according to the linear transmission technique. The present setup is an improvement [15] over the original apparatus [16,17]. A schematic of the present setup is shown in Fig. 1. The electron gun consists of a thorium tungsten filament, an immersion object, an Einzel lens, and defecting plates. The five-cylinder lens consists of five cylindrically symmetric elements with two diaphragms. The data used in the design for the five-cylinder lens are from the tables of Harting and Read [18]. By means of the deflecting plates in front of Faraday cup FC1, the beam can be entirely collected by FC1, thus the primary beams entering the collision chamber (CC) can be recorded by FC1. The geometric length of the CC is 202 mm. The entrance aperture and exit aperture of the CC are 1.2 mm and 1.8 mm, respectively. The exit aperture of the second Fara-

FIG. 1. Schematic of the experimental setup. 1, electron gun; 2, five-cylinder lens; 3 and 8, deflecting plates; 4, FC1; 5, CC; 6, gas inlet; 7, baratron sensor head; 9, FC2; 10, analyzer; 11,Einzel lens; 12, FC3; 13, vacuum connector; 14, valve; 15, retarding plate.

day cup (FC2) is 1.65 mm in diameter and lies 184 mm away from the center of the CC. The solid angle subtended by the entrance aperture of analyzer as seen from the center of the CC was 6.3×10^{-5} sr. The analyzer consists of a five-electrode filter lens and Einzel lens. The transmission beam intensity was collected by the third Faraday cup (FC3) and read out by a D-88 microgalvanometer. The pressure in the CC was measured by an absolute capacitance manometer (mks baratron 127A-113A). The CC and the whole electron-optics system are shielded from Earth's magnetic field and stray magnetic fields by two layers of μ -metal. The butterfly valve under the CC is used to alternate the conditions of the pressure in the CC.

III. PROCEDURE

The principle of the measurement for TCSES (σ ,) using a transmission technique is based on the following law:

$$
\sigma_t = (NL)^{-1} \ln(I_0/I_c) \tag{1}
$$

where N is the atomic density obtained from the temperature inside the CC and the gas pressure in the CC, L is the interaction-region length when a beam passes through the gas, and (I_0/I_c) is the ratio of the beam intensity in front of and behind the CC. Equation (1) is not strictly valid for making the actual measurement because of the small-angle scattering. By extrapolating the elastic differential cross sections experimentally determined by Fink, Jost, and Hermann [19], DuBois and Rudd [20], and Jansen et al. [21], we estimated the maximum relative contributions of the elastic small-angle scattering to TCSES to be less than 0.25% for a C_2H_2 and 0.15% for CO in the electron energy range up to 2600 eV, so that we could neglect it. Inelastic scattering cannot pass through the analyzer because of the action of the retarding field in the analyzer. The energy resolution of the analyzer was about 0.85 eV [15]. Neglecting the scattering, we can determine the TCSES according to Eq. (1). In order to minimize experimental error, the measurements of (I_0/I_c) were performed in alternatively vacuum and gas feed CC conditions. Then Eq. (1) is replaced by Eq. (2):

$$
\sigma_t = -(NL)^{-1} \ln[(I_c/I_0)_g / (I_c/I_0)_v], \qquad (2)
$$

where $(I_c/I_0)_{g}$ and $(I_c/I_0)_{v}$ are the ratios of the transmission-beam intensity to primary-beam intensity with and without gas feed in the CC. The (I_c/I_0) was determined at the evacuated chamber pressure (about 1×10^{-5} Torr) before and after every two sets (i.e., two pressure points). Eight sets were performed at each energy. Considering the space-charge effect, the measuringbeam intensity was kept less than 10 nA. And considering the accuracy of the current meter, the multiple scattering of the beam, and the actual differential pumping conditions, the range of the measuring pressure value was chosen from 0.023 to 1.12 Pa (for C_2H_2) and from 0.040 to 1.55 Pa (for CO). A typical logarithmic plot of transmitted electron-beam intensity against pressure (for C_2H_2) is shown in Fig. 2. The experimental points lie in a straight line, the slope of which gives the TCSES by the least-squares fitting. The temperature inside the CC was estimated by the temperature outside the CC. Since the temperature range outside the CC was between 292 and 306 K and the mks sensor head was maintained at 318.15 K, the pressure in the CC was expected to be slightly lower than the pressure indicated by the mks capacitance manometer, due to the thermal transpiration effect. Blaauw et al. determined experimentally the actual pressure in the CC using the modified Knudsen formula [22]. Poulter et al. [23] have shown that applicability of the Knudsen formula for a given pressure range depends strongly on the kind of gas, and for the correction of pressure due to the thermal transpiration effect, the following semiempirical formula of Takaishi et al. [24] is particularly useful:

$$
\frac{P_M}{P_c} = \frac{AX^2 + BX + C\sqrt{X} + \sqrt{T_M/T_c}}{AX^2 + BX + C\sqrt{X} + 1} \tag{3}
$$

where P_c is the actual pressure, P_M is the pressure indicated by the mks baratron, T_c is the temperature inside the CC, T_M is the temperature in the mks sensor head (here, 318.5 K), and A, B, C , and X are defined in the following equations:

$$
A = A^*(T^*)^{-2}, \t\t(4a)
$$

$$
B = B^*(T^*)^{-1} , \t\t(4b)
$$

$$
C = C^*(T^*)^{-0.5}, \qquad (4c)
$$

$$
T^* = 0.5(T_c + T_M), \qquad (4d)
$$

$$
X = 0.133 \, P_M d \, , \tag{4e}
$$

where d is the diameter (in m) of the connector between the mks sensor head and the CC (here, $d=0.0076$ m), and the units of pressure are all in Pa. The values of the reduced constants A^* , B^* , C^* , which depend on the molecular diameter, were calculated according the relations of Takaishi et al. (see Ref. [23]). Replacing P_M

FIG. 2. The ratio of transmitted-beam intensity vs pressure in the CC at selected energies from 400 to 2400 eV for C_2H_2 .

with P_c calculated by Eq. (3), this correction for TCSES resulted in an increase of TCSES on average, and it was estimated to be about 2.2% for C_2H_2 and 1.8% for CO. The accuracy of the pressure measurements was estimated as better than 1% according to the baratron manual and the measuring conditions. The mks baratron measurement had been compared with a McLeod gauge measurement within the deviation, which is less than 3% . Considering the actual differential pumping conditions, the interaction-region length (L) can be determined by expression

$$
L = L_g + d_1 + d_2 + L_p \t\t(5)
$$

where L_g is the geometric length of the CO, d_1 and d_2 are the diameters of the orifices at the both ends of the CC, and L_p is the estimated value depending on the differential pumping conditions at that time (less than 10 mm). Determination of the electron impact energies was performed by means of the 6(1/2) digital voltmeter. The purities are 99.99% for CO and 99.9% for C_2H_2 .

IV. RESULTS, ERRORS, AND DISCUSSION

Our experimental results of TCSES for CO and C_2H_2 are shown in Table I and Figs. 3 and 4, along with the existing experimental data, and theoretical data in the energy range 381—3000 eV is shown in Fig. 3. Each entry of our results given in Table I is the weighted average of mostly from two to four independent experimental runs. The estimated total experimental error of our results is about 5%, including the systematic error of $1\% - 3\%$ and the statistical error of 2.5%—3%. The errors of our results are given in Table I and shown in Figs. 3 and 4. The systematic errors mainly result from two factors: the decrease of $(I_c/I_0)_v$, especially at both ends of the electron energy range measured, and the increasing inaccuracy, frequently at the high end of the pressure range being

TABLE I. Total cross sections for electron scattering (in 10^{-20} m²). The total errors (in %) are given in parentheses.

Energy (eV)	$_{\rm CO}$	C_2H_2
400	4.20(5.5)	5.38 (5.7)
500	3.61(5.3)	4.59 (5.5)
600	3.16(5.5)	4.06(5.4)
700	2.88(4.7)	3.67(5.0)
800	2.55(4.5)	3.19(4.7)
900	2.37(4.6)	2.86(4.5)
1000	2.18(4.0)	2.61(4.5)
1100	2.06(3.7)	2.41(4.5)
1200	1.92(3.8)	2.22(4.5)
1300	1.73(3.5)	2.11(4.5)
1400	1.70(3.0)	1.95(4.5)
1500	1.52(3.5)	1.84(4.5)
1600	1.50(3.7)	1.74(4.7)
1800	1.34(3.5)	1.57(4.7)
2000	1.26(4.0)	1.44(4.5)
2200	1.15(4.5)	1.36(4.3)
2400	1.06(4.3)	1.32(5.0)
2600	1.02(4.8)	1.20(5.5)

FIG. 3. Total cross sections for electron-CO scattering. Present results are compared with the results of Karwasz et al. (Ref. [8]), Garcia, Aragon, and Campos (Ref. [9]), Kwan et al. (Ref. [5]), Sueoka and Mori (Ref. [6]), and the theoretic data of Jain and Baluja (Ref. [14]). \circ , present results; A, Ref. [8]; \times , Ref. [9]; \blacksquare , Ref. [5]; \blacklozenge , Ref. [6]; ∇ , Ref. [14].

measured, when plotting the logarithmic values of the transmitted electron-beam intensity against pressures. The first factor is a result of the beam conditions going bad and the second factor is attributable to the multiple scattering due to the insufficiency of the differential pumping. The other systematic errors produced by the correction of pressure according Eq. (3) and resulting in the measured differences among individuals are all less than 1%. The statistical errors are estimated as follows: errors caused by instability of gas feed and power supply are about 1.5%; errors produced by observational error and inability to determine the interaction-region length are all about 1%; errors in the measurement of pressure by means of the baratron and in the determination of the temperature in the CC are all less than 1%; errors made in the correction of pressure for the thermal transpiration effect was estimated to be less than 0.5%; errors resulting

FIG. 4. Total cross sections for electron-C₂H₂ scattering. \circ , this work; $+$, Sueoka and Mori (Ref. [11]); \blacktriangledown , Jain and Baluja (Ref. [14]).

from the uncertainty of the electron energy definition and from the nonlinearity of the current meter are all less than 0.5%.

Comparing the TCSES of CO in Fig. 3, the results of Garcia, Aragon, and Campos [9] with our results within experimental error in the overlapping energy range; the results of Karwasz et al. [8] agree with our results within experimental error in the energy range 400—1000 eV and are lower than our data above 1000 eV (lower by 12% at 2000 eV and lower by 15% at 2500 eV). This discrepancy was caused by the systematic errors between different setups, which may result from forward-scattering contamination and multiple-scattering effect as discussed by Ma, Liescheski, and Bonham [26]. The data of Kwan et al. [5] at 400 and 500 eV also agree with our results. That of Sueoka and Mori $[6]$ is lower by 13% than ours (lower by 12% than those of Garcia, Aragon, and Campos [9] at 400 eV). For C_2H_2 the result of Sueoka and Mori [11] is lower by 9% at 400 eV than ours. We noticed that the TCSES of Sueoka and Mori on N_2 [6] and CH₄ molecules [25] are all lower than existing data in the overlapping

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energy range. No data for C_2H_2 can be compared above 400 eV with ours. The data of CO calculated by Jain and Baluja [14] are higher by 19%, 12%, and 5% at 500, 1000, and 2000 eV, respectively, than ours. For C_2H_2 the data of Jain and Baluja [14] are higher by 7% at 500 eV and higher by 4% at 1000 eV than our data, and at 2000 eV with ours within statistical error.

From our results, the TCSES for C_2H_2 , compared to those of CO at the same energy, are higher by about 20% within statistical error. Our results also show the similarities in the energy dependence of TCSES for isoelectronic molecules.

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