Remarks on total and elastic cross sections for electron and positron scattering from CO_2

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There remain discrepancies in the determination of the total and elastic cross sections for electron and positron collisions with carbon dioxide at impact energies from 0.3 to 100 eV. We have carried out a joint experimental and theoretical study in an attempt to resolve the differences. Our measurements for total cross sections for electron impact agree extremely well with those of Hoffman *et al.* [Phys. Rev. A **25**, 1393 (1982)] above 2 eV, while those for positron impact agree to better than 15% in magnitude, and are in an excellent accord for the energy-dependence. The present observations, along with an analysis using a knowledge of the dominant inelastic processes, lead us to conclude that the total cross section determined in earlier studies is a considerable underestimate at intermediate energies, where inelastic processes are not negligible. The analysis provides a rationale for understanding the discrepancies. Furthermore, we present that the total cross section by positron impact below 2 eV becomes larger by 20% than that of electron impact, and provide a rationale as well as implication to a possible application. © *1997 American Institute of Physics.* [S0021-9606(97)01841-2]

I. INTRODUCTION

Due to their fundamental nature as well as applications from astrophysics and aeronomy to plasma chemistry, the study of the total, elastic and inelastic processes in electron scattering from carbon dioxide (CO_2) remains important.[1] Itikawa and Shimizu[2] have compiled a set of cross sections of electron scattering from CO_2 for a variety of elastic and inelastic processes in a wide range of collision energies by collecting all the published data and critically examining them. The data they reported have been widely used in applications, particularly to astrophysics and radiation physics.

Since then, there have been numerous experimental studies[3–10] based on electron beam and swarm measurements and theoretical studies[11–16] based on close coupling methods (using different forms of polarization and exchange potentials). These studies have led to considerable improvement in our understanding of electron scattering by the molecule. There remain issues to be resolved corresponding processes in CO_2 scattering, and even for total and elastic scattering, agreement among the various experiments and between theory and experiment is not yet satisfactory for practical purposes, especially at energies below 100 eV. However, we believe that more reliable results have been reported in the total and elastic cross sections,[17] and a more realistic cross-section data set can now be constructed.

Positron-scattering experiments have been carried out by Hoffman *et al.*,[10] Sueoka and Hamada,[18] Charlton *et al.*,[19] and Horbatsch and Darewych[20] have given a theoretical analysis. In theory, electron/positron interactions with the target molecule are approximated by the static, exchange and correlation-polarization terms. For electron impact, the static and correlation-polarization interactions are both attractive, and the exchange interaction is known to play an important role when the incident electron energy is low. These three interactions contribute in a rather complex manner to the scattering process. For positron scattering, the static interaction is repulsive, while the correlation-polarization interaction is attractive, and no exchange interaction exists, and hence, the repulsive static and attractive polarization terms cancel out, resulting much small effect on the scattering at low-to-intermediate scattering energies. As the incident energy increases above 100 eV, interactions become more impulsive and Born-type perturbative approaches are valid and frequently used for describing scattering dynamics, leading to a similar magnitude of the cross sections for both projectiles. By studying these two-projectile cases comparatively, we may understand better the details of interaction and the underlying dynamics. For positron impact, the results of Hoffman *et al.*[10] and Sueoka and Hamada[18] have suggested there might be a region where the total cross section by pos-

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itron impact is larger than that by electron impact. If so, there may be a situation in which positron impact produces more fragments of a target molecule for a specific process in a certain energy region or more vibrational and rotational excitations, which may well be important in application.

In this short note, we report our evaluation of the total cross section based both on our new measurements for total (0.7–600 eV, Sueoka and Hamada both by electron and positron impacts) and elastic (1.5–100 eV, Tanaka by electron impact) cross sections and on theoretical (2.0–100 eV, Kimura, Takekawa, and Itikawa) calculations for elastic collisions, along with earlier published results. We identify the cause of discrepancies, and we set realistic upper and lower limits to the total cross section. We also comment on the contributions of the rovibrational excitation channels to total cross section. We re-examine the findings by Hoffman *et al.*, and Sueoka and Hamada that the positron impact cross section is larger than that of electron impact below 2 eV, and we provide a rationale.

The detailed experimental procedures and theoretical models we have used have been described elsewhere.[17,18] For electron and positron scattering, the present experiment concentrates on an accurate measurement in the higher energy region above 2 eV, while the previous measurement[18] was for lower-energies below 2 eV. Both agree well in the matching energy region to within 10%. The data presented here are a combination of the two measurements.

The threshold energies for vibrational and electronic excitation channels are[21]

$$\text{vibrational excitation:} \quad (100) = 0.1653 \text{ eV},$$
$$(010) = 0.0827 \text{ eV},$$
$$(001) = 0.2912 \text{ eV}.$$

For electronically excited states, little is known except for some Rydberg states.[22] However, some low-lying optically allowed excited states have been studied with the thresholds,

$$\text{electronic excitation:} \quad {}^1\Sigma_u = 11.04 - 11.05 \text{ eV},$$
$${}^1\Pi_u = 11.38 - 11.40 \text{ eV}.$$

In addition, experiments based on photoabsorption have assigned a few singlet excited states between 7–10 eV along with low-lying triplet excited states with threshold energies around 6 eV, but exact energy values and their feature are still uncertain.

First, we discuss the results for electron scattering, and then the results for positron scattering. We divide our discussion into three specific scattering-energy regions, namely, (a) higher energy region above $E > 7$ eV where an electronic excitation channel opens, (b) intermediate energy region which covers a prominent resonance peak ($2 \text{ eV} < E < 7 \text{ eV}$, the dominant inelastic processes are vibrational excitation in this energy region), and (c) low energy region below 2 eV. Lastly, we compare electron- and positron-scattering.

In this approach, we contrast the agreement and deviation among experiments and theories, and suggest a recommended set of the total cross section data.

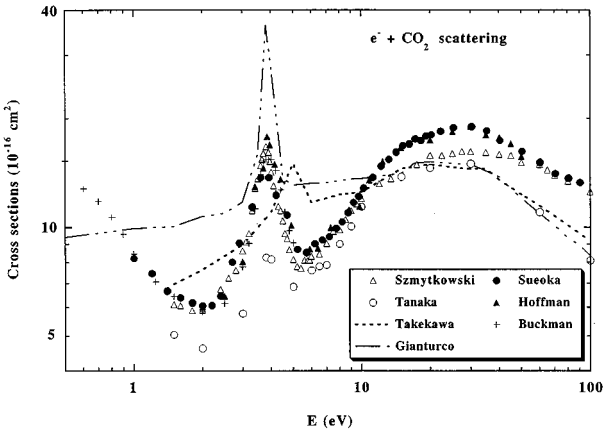

FIG. 1. Total and elastic cross sections for electron scattering. The data shown are indicated in the figure. The data labeled as Sueoka are the present measurements for total cross sections, while those labeled as Tanaka are the present elastic cross sections. The data labeled as Szmytkowski, Hoffman, and Buckman are all for total cross section measurements. Those labeled by Takekawa are the present theory for elastic collisions, while those by Gianturco are other theoretical elastic results described in detail in the text.

II. ELECTRON IMPACT

Figure 1 shows the total and elastic cross sections for electron impacts which we believed to be reliable, based on experimental and theoretical studies.

Note that the data labeled as Sueoka are the present measurement for total cross sections, while those labeled as Tanaka are the present elastic cross sections. The data labeled as Szmytkowski, Hoffman, and Buckman are the total cross section measurements. Those labeled by Takekawa are the present theory for elastic collisions, while those by Gianturco are other theoretical elastic results described in detail below.

A. High energy region

The present total cross section agrees closely with the data of Hoffman *et al.*[10] in magnitude and energy-dependence of all energies. The total cross-section data by Szmytkowski *et al.*[8] are in a good accord with the present data and with Hoffman *et al.* below 10 eV, but become smaller by at least 15% above 10 eV–50 eV. Above 50 eV, they again approach and agree with the present results. Interestingly, the data by Szmytkowski *et al.* agree reasonably well with those by Tanaka *et al.*[17] who measured elastic cross section up to 100 eV. The present theoretical results for elastic scattering (see Ref. 15 for more details) as well as those of Gianturco *et al.*[16] agree excellently with elastic cross section measurements by Tanaka *et al.* above 12 eV.[17] It is expected that the theoretical elastic results become smaller than the experimental total cross sections above 10 eV. Below 10 eV, the theoretical results begin to deviate. The origin of this deviation is discussed below. According to Nakamura[7] who determined cross sections for all CO_2 processes based on swarm measurements, electronic excitation cross sections (threshold >7 eV) begin to increase rapidly as soon as they pass the threshold energies and reach values as large as 10^{-16} cm^2 at 15 eV. In his analysis, Nakamura

adopted a ''two-band'' approximation to represent all electronic excitation channels with thresholds of 7.8 eV and 10.5 eV, respectively, and utilized the swarm data by Hake and Phelps.[23] (Nakamura assumed that vibrational excitation drops sharply in this energy domain except for the antisymmetric stretching mode. However, this assumption may be questionable.) The swarm measurement is known to provide somewhat ambiguous results in the energy region above a few eV. If we allow about 50% correction to these electronic excitations, the sum of these electronic excitation cross sections and the elastic cross section of Tanaka *et al.* agrees well with the present total results as well as those of Hoffman *et al.* Therefore, we believe that the total cross section determined by Szmytkowski *et al.* underestimates by at least 15% in their energy region. The good agreement of the present results for theoretical elastic scattering also supports the reasonable accuracy in the measured elastic cross section.

B. Intermediate energy region

The present total measurements again agree very well with those of Hoffman *et al.* in this energy region, and give the nearly identical peak position of the resonance. However, the present peak height is smaller by about 25%–30%. The result by Buckman *et al.*[24] is in excellent accord with the present data and those of Hoffman *et al.* although the peak height of the 3.8 eV-resonance by Buckman *et al.* lies between ours and that of Hoffman *et al.* The data by Szmytkowski are consistently smaller by 5%–10% at all energies except for the peak where Szmytkowski's data are in a good agreement with those of Hoffman *et al.* Elastic data by Tanaka *et al.* are smaller by 10%–15% from the present results and by 8% from those of Szmytkowski's at 5–7 eV, while the difference grows to a factor of 2–3 at the peak position of the resonance. For the peak value, if we take the vibrational excitation cross section by Register *et al.*,[4] adding to the elastic data by Tanaka *et al.*, then the total value comes out very close to that by Hoffman *et al.*, and by Szmytkowski *et al.*, and therefore, the present peak value in the total may be a slight underestimate. As is well known, the 3.8 eV-resonance arises from strong vibrational excitation[25] through the shape resonance and hence, a large contribution from these vibrational excitation channels to total is expected. Another set of total cross section measurement by Strakeljahn *et al.*[26] exists in this domain (not shown). Their results seem to be in a good harmony with that of Buckman *et al.* although the result is somewhat smaller by a few %. The present theoretical result is unable to reproduce the peak position and height since doing so is known to require a careful selection of polarization and exchange potentials in the calculation. The theoretical result by Gianturco *et al.*[16] is able to reproduce the peak position, but the magnitude is larger by a factor of 2. The first electronic excitation of a $^3\Sigma$ state (the threshold energy 5.4 eV) begins at the higher end of the resonance peak, and this channel is expected to make some contribution to the total. The study of this excitation channel is underway in this group.

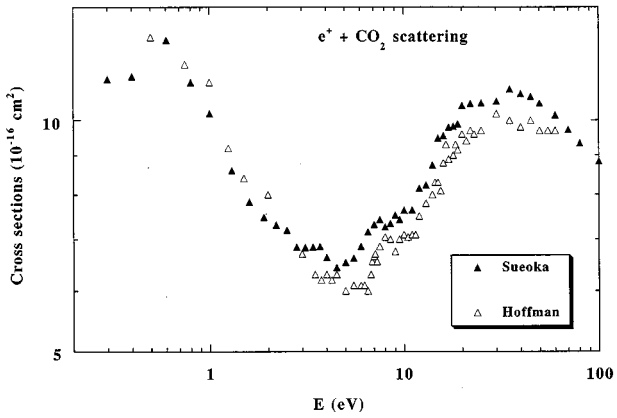

FIG. 2. Total cross sections for positron scattering. The data shown are indicated in the figure. The data labeled as Sueoka are the present measurements, while those of Hoffman are other measurements.

C. Low energy region

In this domain, the present measurement enjoys excellent agreement with Buckman *et al.*[24] Vibrational excitation channels are dominant and strong contributors to inelastic processes down to their thresholds. A large difference between elastic and total cross sections is expected. A simple extrapolation of elastic data by Tanaka *et al.* below 1.5 eV suggests that the difference between the two is approximately 20%–25%, and as described, the vibrational excitation may be able to account for all the rest. By taking the vibrational excitation data from Ref. 23 and summing with the elastic by Tanaka *et al.* the total cross section obtained is indeed nearly identical to the present result. Other data in this region are the theoretical results by Gianturco *et al.*, which are nearly independent of the scattering energy with a slowly decreasing trend toward zero energy. This trend contradicts the trends of the present result as well as those of Buckmann *et al.*, which show an rapid increase below 2 eV. The swarm data by Nakamura for momentum transfer cross section show a strong increasing trend, consistent with the present result. Because of the complexity of the problem in accurately determining the correlation-polarization potential, the *ab initio* approach may not offer an accurate result in this energy region. In the low-energy limit, according to the result by Buchman *et al.*, the total cross section continues to increase as the scattering energy decreases, and at the zero energy limit, it should approach a finite value of the cross section based on scattering length arguments.

III. POSITRON IMPACT

Figure 2 displays the total cross sections obtained with the present measurements and by Hoffman *et al.* The data below 2 eV are from our earlier study.[18]

Note that the data labeled as Sueoka are the present measurement, while those of Hoffman are other measurements.

A. High energy region

The present results and those of Hoffman *et al.* show a good accord in energy-dependence, but the magnitude of the

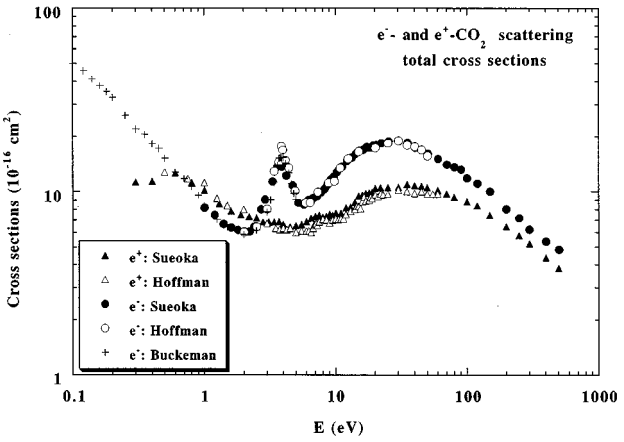

FIG. 3. Comparison in the total cross sections by electron and positron scattering. We include only the present result, Hoffman *et al.* (Ref. 10) and Buckman *et al.* (Ref. 24) for electron scattering as noted in the figure.

present results is higher by a few %. Both show small oscillatory structures; a small hump at 15 eV corresponds to the opening of the CO_2 ionization channels, and that at near 7.8 eV to positronium formation. Above 200 eV or so, the present positron scattering and electron scattering cross sections begin to merge, indicating entrance to the perturbative domain. A large structure at 30 eV is interesting, and we speculate that the origin of this hump may be due to an opening of another ionization channel.

B. Intermediate energy region

The present results and those of Hoffman *et al.* agree quite well in this energy domain, although, again, the present results are somewhat larger in magnitude. The strong resonance peak seen in electron impact at 3.8 eV is *not* present in positron impact, suggesting a different interaction mechanism and energy-dependence between incoming positron- and electron-nuclear interactions. Positron scattering cross sections gradually increase below 5 eV. This increasing trend is similar qualitatively to that of electron scattering below 2.5 eV (see Fig. 3). From a theoretical point of view, there is no reason for electron and positron scattering cross sections to behave similarly on this energy region, and hence, this similarity may be accidental in the sense that three interaction terms in electron scattering and two interaction terms in positron scattering coincide in this energy region.

C. Low energy region

The two results show good agreement in this region. The present cross sections appear to be slightly smaller by a few % than those by Hoffman *et al.* below 2 eV, but both increase rather rapidly below 1.5 eV. Below 0.7 eV, the present result levels off, giving a maximum value at 0.6 eV, and then follow a slow decrease. The maximum value is approximately 13×10^{-16} cm^2 which appears as a broad peak at this scattering energy. The data by Charlton *et al.*[19] (not shown) are somewhat smaller than ours and Hoffman's at all energies, but show also a slight increasing trend below 2 eV. In order to examine the origin of this broad peak above, it will be extremely interesting to carry out further detailed experimental study for much lower energies.

IV. COMPARISON BETWEEN ELECTRON SCATTERING AND POSITRON SCATTERING

Figure 3 is a comparison of cross sections by the electron and positron impacts for the present study (lebeled as Sueoka) and for Hoffman *et al.* A few characteristic features should be noted: (i) below 2.5 eV, the magnitude of the total cross section by positron impact becomes larger than that by electron impact. This was speculated experimentally by Hoffman *et al.* earlier, but they gave no firm data or corresponding interpretation. This reverse of two cross sections in magnitude has been observed for other systems[27] such as NH_3, and may be a case in which some modes of rovibrational excitations become larger for the positron scattering compared to those in electron scattering at certain energies. This larger cross sections by positron impact may be significant for applications. If the positron impact can excite a certain type of a rovibrational mode or electronic state, this may lead to specific fragmentation products or excited species. Then, the positron impact technique could be usable for a selective production of specific fragments or radicals of molecules, (ii) resonances (shape or Feshbach) are common features in electron-molecule scattering. The 3.8 eV resonance peak in electron scattering (shape resonance) is not present in the positron scattering. This suggests a different energy dependence for the interaction between an electron and positron with the target nuclear motion. It is practically important to search for any type of resonances in the positron scattering as a function of collision energy in various scattering processes. But little is known about resonances in positron-molecule scattering. The structure seen here could be due to some type of resonance. Particularly, when an incomig positron may be temporarily trapped by the polarized electron-cloud of the target molecule, i.e., ''positronium in a molecule,'' then this may lead to a resonance in elastic process. Further detailed experimental and theoretical studies on resonances in the positron scattering are needed, (iii) the cross sections by electron and positron impact which appear to cross over each other around 0.6–0.7 eV [see (i) above] reverse the order with a larger value for electron scattering at much lower energies. The cancellation of the static and correlation-polarization potential may be responsible, and (iv) at zero-energy limit, the two cross sections approach different limits. The knowledge of the exact energy region where these two cross sections begin to diverge would provide a more accurate testing ground for interaction potentials.

The behavior of rotational excitation cross sections near threshold is known to be described by the Born approximation reasonably well, which suggests that when a molecule has a quadrupole moment with a negative value, the rotational excitation cross section by the positron impact should become larger than that by electron impact.[28] This may in

fact be responsible to the reverse of the cross section described in (i) above.

In summary, we present accurate total cross sections by the electron and positron scattering from the CO_2 molecule. The total cross section determined by Szmytkowski *et al.* underestimates at least 15% at all energies. A larger deviation at intermediate energies occurs. The present study of positron scattering shows the energy region where the positron total cross section becomes larger than that by electron scattering. This observation might have a potential significance for application, because selectively, we may be able to generate specific fragment products or rovibrational excitation species by using positron impact.

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