Electron scattering cross sections for momentum transfer and inelastic excitation in carbon monoxide[a)]

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Cross sections for momentum transfer and for rotational, vibrational, and electronic excitation were obtained from an analysis of experimental data for electron drift velocity, characteristic energy, and ionization coefficient. Recently measured data in the range $1\times10^{-16}\leq E/N\leq 4\times10^{-15}$ V cm^2 were included in the analysis.

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I. INTRODUCTION

Cross sections for low-energy electron-CO scattering are required for modeling plasmas containing CO, as in gas-discharge lasers and in coal-fired MHD generators. Normalized low-energy electron scattering data are customarily obtained through the analysis of measured electron transport coefficient data.[1-5] Using this method, initial information on the CO momentum-transfer cross section at thermal energies was obtained by Pack, Voshall, and Phelps,[6] and cross sections for momentum transfer, rotational excitation, and direct vibrational excitation were obtained by Hake and Phelps.[7] The electron-swarm data then available restricted the analysis to electron energies less than 1 eV. Recent experimental data at higher energies provide previously unavailable information on the electron drift velocity[8,9] w and the electron characteristic energy[10] $\epsilon_k=eD/\mu$ (where D is the coefficient for electron diffusion in a direction perpendicular to the applied electron field E, and $\mu=w/E$ is the electron mobility). Some of these recent data were used by Saelee and Lucas[9] in a Monte Carlo simulation to provide information on the excitation of electronic states in CO.

The present analysis is based on these recent experimental electron transport coefficient data.[8-10] The momentum-transfer cross section is revised, and information is provided on rotational, vibrational, and electronic excitation. Some of these results were reported previously in preliminary form.[11]

II. METHOD

In the method of swarm analysis,[1-5] cross sections for momentum transfer, and for rotational, vibrational, and electronic excitation, are found by manually adjusting the cross-section data for those processes until values of the transport coefficients calculated from the cross-section set agree with experimentally measured values. The results of electron-beam experiments and of theory are included in the analysis wherever possible. In cases where accurate experimental data are available, this method has yielded cross sections with high precision, although with limited fractional energy resolution. The present analysis used a computer program developed by Phelps and associates[1,2,12] which numerically integrates the Boltzmann equation for the electron distribution function, from which the electron transport coefficients are calculated. The theoretical basis for these computations is the spherical harmonic expansion approximation.[13]

The electron drift velocity data used in this analysis (Fig. 1) are the measurements of Pack *et al.*[6] at 77 K from E/N (ratio of applied electric field to gas density) $=1\times10^{-19}$ to 1×10^{-17} V cm^2 and at 300 K from $E/N=1\times10^{-17}$ to 1×10^{-16}, recent data of Milloy[8] at 294 K from $E/N=1\times10^{-16}$ to 1×10^{-15}, and recent data of Saelee and Lucas[9] from $E/N=1\times10^{-15}$ to 5.7×10^{-15}. The characteristic energy data (Fig. 1) are from Warren and Parker[14] at 77 K from $E/N=1\times10^{-19}$ to 1.8×10^{-16} V cm^2, from Skinker and White[15] at 288 K from $E/N=1.8\times10^{-16}$ to 1.6×10^{-15}, and recent data from Lakshminarasimha *et al.*[10] from $E/N=1\times10^{-15}$ to 1.4×10^{-14}. As previously observed,[7] a good separation of elastic and inelastic effects in CO is obtained by using collision frequencies[1,2] (Fig. 2) calculated from the experimental w and ϵ_k data. The momentum-trans-

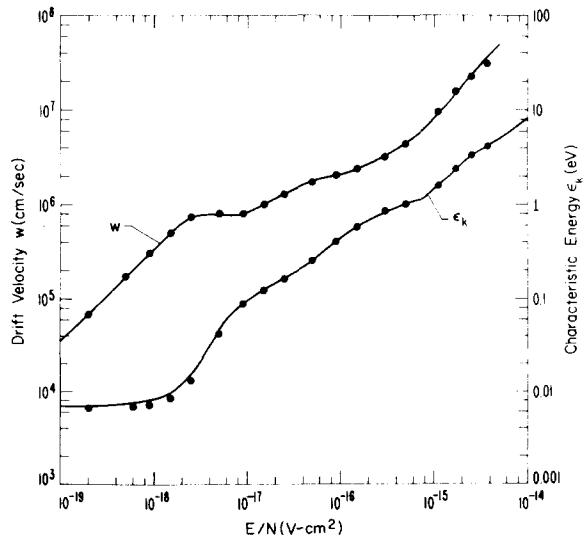

FIG. 1. Drift velocity w and characteristic energy ϵ_k of electrons in CO as a function of E/N. The solid curves show the experimentally measured data, and the points were calculated using the cross sections determined in this analysis.

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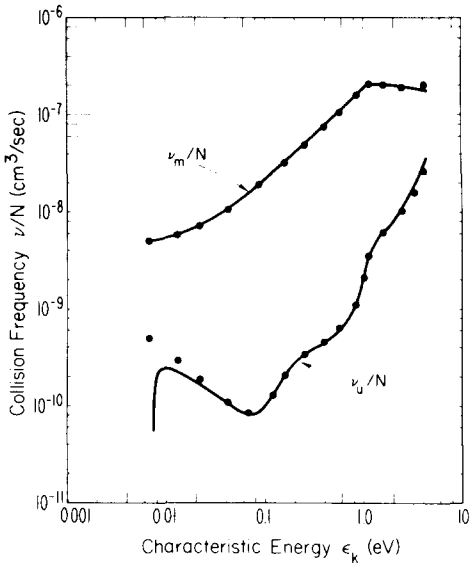

FIG. 2. Collision frequencies for momentum transfer ν_m/N and for energy exchange ν_u/N for electrons in CO as a function of characteristic energy ϵ_k. The solid curves were calculated from the experimental data of Fig. 1. The points were calculated using the cross sections determined in this analysis. The fit of the ν_u/N data near 0.01 eV is discussed in Ref. 6.

fer collision frequency, $\nu_m/N=(e/m)E/Nw$, is affected mainly by the momentum-transfer cross section; the collision frequency for energy exchange, $\nu_u/N=ew(E/N)/(\epsilon_k-kT)$, is affected mainly by the inelastic cross sections. At the highest values of E/N considered here (1×10^{-15} to 4×10^{-15} V cm^2), the ionization coefficient measurements of Bhalla and Craggs[16] and of Lakshminarasimha *et al.*[10] were also used (Fig. 3). Much of the above swarm data are available in tabular form in the review by Dutton.[17]

III. RESULTS

A. Momentum-transfer cross section

The cross section for momentum transfer is defined as

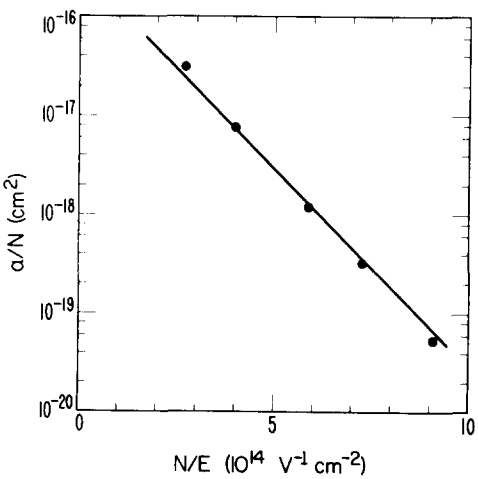

FIG. 3. Ionization coefficient α/N for electrons in CO as a function of N/E. The solid curve shows experimentally measured data. The points were calculated using the final cross sections determined in this analysis.

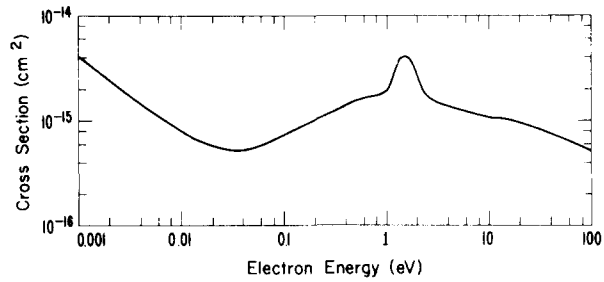

FIG. 4. The momentum-transfer cross section Q_m for electrons in CO determined in the present analysis, as a function of electron energy. Values are given in Table I.

$$Q_m=\int(1-\cos\theta)\frac{d\sigma}{d\Omega}\,d\Omega, \tag{1}$$

where θ is the electron scattering angle and $d\sigma/d\Omega$ is the total absolute differential cross section for electron scattering. The present Q_m results (Fig. 4 and Table I) fall into three energy ranges. For energies less than 0.3 eV, we find that the Q_m values previously determined by Hake and Phelps[7] are consistent with all experimental swarm measurements to date. These values agree within $\pm10\%$ with the theoretical calculations of Crawford and Dalgarno.[18] For energies from 0.3 to 3 eV, where the recent experimental swarm data provide new information, it was necessary to increase the Hake-Phelps cross section by 7–25% for consistency with the experimental swarm data. At energies greater than 3 eV, we have given Q_m a negative slope as discussed in Sec. III D.

The values of ν_m/N calculated from this revised momentum-transfer cross section are shown in Fig. 2. Agreement with the experimental data is very good, well within the accuracy of the measured transport coefficient data (which we estimate to be $\pm5\%$, except at the highest and lowest values of E/N where the experimental data are somewhat

TABLE I. Momentum-transfer cross section for CO as a function of electron energy, as determined in the present analysis. The values correspond to the curve in Fig. 4.

E (eV)	Q_m (10^{-16} cm^2)	E (eV)	Q_m (10^{-16} cm^2)	E (eV)	Q_m (10^{-16} cm^2)
0.0	60.0	0.25	11.2	3.3	14.6
0.0010	40.0	0.30	12.1	3.6	14.2
0.0020	25.0	0.35	13.0	4.0	13.8
0.0030	17.7	0.40	13.8	4.5	13.3
0.0050	12.3	0.50	15.4	5.0	12.9
0.0070	9.8	0.70	16.5	6.0	12.3
0.0085	8.6	1.00	18.5	7.0	11.8
0.0100	7.8	1.2	28.0	8.0	11.3
0.015	6.5	1.3	37.0	10.0	10.6
0.020	5.9	1.5	42.0	12.0	10.4
0.030	5.4	1.7	40.0	15.0	10.2
0.040	5.2	1.9	32.0	17.0	10.1
0.050	5.4	2.1	23.5	20.0	9.8
0.070	6.1	2.2	21.5	25.0	9.1
0.100	7.3	2.5	17.5	30.0	8.6
0.15	8.8	2.8	16.0	50.0	7.1
0.20	10.0	3.0	15.4	75.0	6.1
				100.0	5.5

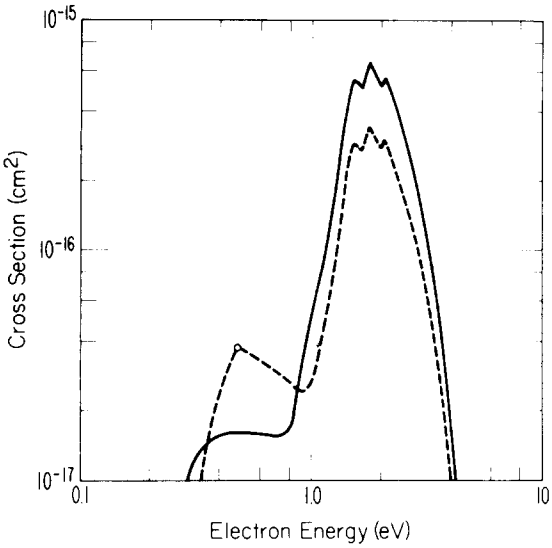

FIG. 5. Cross section for excitation of the $v=1$ level of CO as a function of incident electron energy. The dashed curve for energies greater than 0.48 eV is the experimental data of Ref. 19. The first data point at 0.48 eV is connected with the threshold by the linear curve shown. The solid curve is the cross section determined in the present analysis: For energies from threshold to 0.7 eV, the curve is based on the cross section of Ref. 6; for energies greater than 1 eV, the solid curve is the dashed curve increased by 90%; from 0.7 to 1 eV, the curve smoothly connects the other two portions in a way which provides a fit to ν_u/N data. Values for the solid curve are tabulated in Table II.

less reliable). The Q_m values of Fig. 4 and Table I are estimated to be accurate within $\pm 10\%$ for energies of 3 eV and less, but as the energy increases, the error could increase to as large as $\pm 30\%$.

B. Rotational excitation

The analysis of Hake and Phelps[7] included rotational excitation due to the permanent dipole moment of CO. Those authors obtained satisfactory agreement with experimental transport coefficient data, but suggested that even better agreement might result from including excitation due to the quadrupole moment. This point has been discussed in relation to theoretical calculations of rotational excitation in CO.[18-20] The present analysis included rotational excitation due to both dipole and quadrupole moments in the continuous approximation.[1,7] This approximation does not include the resonant structure in the rotational cross section near 1.7 eV; however, the error incurred in omitting the resonance is expected to be small, since the energy lost to rotational excitation in the resonance region is many times smaller than that due to vibrational excitation. The dipole moment was fixed at the value determined in Stark-shift measurements,[21] $0.044ea_0=0.112\times10^{-18}$ esu cm, and the quadrupole moment was varied to obtain the best fit to ν_u/N data near $\epsilon_k=0.07$ eV (Fig. 2). The optimum fit, for a quadrupole moment of $(1.34\pm0.15)ea_0^2=(1.8\pm0.2)\times10^{-26}$ esu cm^2, is better than that obtained by Hake and Phelps and also better than that using the recommended value[22] of the quadrupole moment, $1.86ea_0^2=2.5\times10^{-26}$ esu cm^2. The disagreement between calculated and experimentally derived ν_u/N data at very low characteristic energies, $\epsilon_k\sim0.01$ eV (Fig. 2), was previously observed and discussed by Hake and Phelps.[7]

C. Vibrational excitation

The electron-impact excitation of ground-state CO ($v=0$) to higher vibrational levels can proceed by one of two processes.[2] At energies less than 1 eV, *direct* excitation takes place due to the permanent dipole moment of CO. As expected from the approximate selection rule[23] $\Delta v=1$, this process is observed only in the $v=1$ cross section. At energies between 1 and 3 eV, *resonant* excitation takes place via the compound negative ion state, CO^-. This resonance is present in all the cross sections, $v=1, 2,...,10$. These two processes will now be discussed in turn.

The $v=1$ cross section for direct excitation, from threshold to 1 eV, has been discussed by Phelps[2] and by Schulz.[23] Briefly, cross sections measured in electron-beam-scattering experiments[24,25] are approximately a factor of 2 larger than the cross section determined in the swarm analysis of Hake and Phelps.[7] This discrepancy was considered in detail in the present analysis, since the recently measured electron transport coefficient data provide new information in this energy range, and since the swarm analysis method is particularly sensitive to the cross section at the threshold of an inelastic process,[2] such as the onset of vibrational excitation.

The electron-beam-scattering results of Ehrhardt,

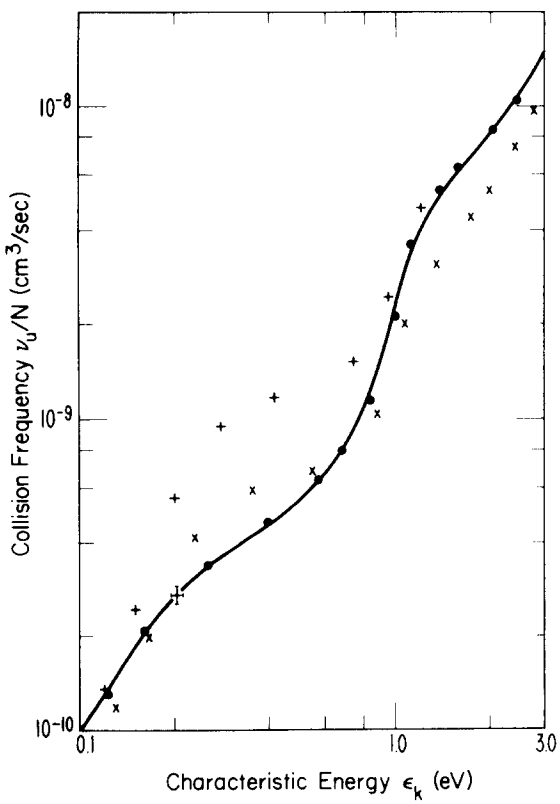

FIG. 6. Expanded plot of collision frequency for energy exchange ν_u/N as a function of characteristic energy ϵ_k. The solid curve was calculated from the experimental data of Fig. 1, and an estimate of the experimental accuracy is shown by error bars. The points were calculated from different cross sections for vibrational excitation: solid points, the cross sections determined in the present analysis (i.e., for $v=1$, the solid curve in Fig. 5); $\times$, cross sections of Ref. 19 (i.e., for $v=1$, the dashed curve in Fig. 5); $+$, cross sections of Ref. 19 increased by 90%.

TABLE II. Cross section for excitation of the $v=1$ level of CO as a function of electron energy, as determined in the present analysis. The values correspond to the solid curve in Fig. 5.

E (eV)	$Q(v=1)$ (10^{-16} cm^2)	E (eV)	$Q(v=1)$ (10^{-16}cm^2)	E (eV)	$Q(v=1)$ (10^{-16} cm^2)
0.266	0.0	1.0	0.513	2.086	5.683
0.290	0.095	1.031	0.604	2.170	4.965
0.32	0.125	1.13	0.924	2.281	4.176
0.35	0.144	1.215	1.350	2.316	4.285
0.40	0.156	1.307	2.137	2.404	3.747
0.50	0.159	1.410	3.602	2.508	3.120
0.60	0.157	1.514	5.377	2.688	2.455
0.70	0.154	1.645	5.054	2.872	1.828
0.80	0.165	1.740	5.934	3.072	1.290
0.85	0.224	1.821	6.578	3.294	0.861
0.90	0.300	1.902	5.843	3.528	0.555
0.95	0.397	1.982	5.216	3.816	0.287

Langhans, Linder, and Taylor[25] (Fig. 5) are similar to those obtained by Schulz.[24] Since the experimental data of Ref. 25 are available only for electron energies of 0.48 eV and greater, it is necessary to connect their data with the $v=1$ threshold at .266 eV. Following Phelps[26] and Schulz,[23] we have chosen a straight line. As discussed by Schulz,[23] this straight line has a slope comparable to (although somewhat larger than) that measured in a trapped-electron experiment by Burrow and Schulz.[27] Using this cross section, our calculated values of ν_u/N differ from the experimental values (Fig. 6) by more than the estimated accuracy of the measured data. When this cross section is increased by 90% (for reasons discussed below) the discrepancy is even greater (Fig. 6). We conclude, as did Schulz,[23] that the cross section measured by Ehrhardt *et al.*[25] (which is similar to the previous measurement of Schulz[24]) is inconsistent with electron transport coefficient data. Part of this discrepancy may be due to different angular distributions for direct and resonant scattering.[26]

The direct excitation cross section from the analysis of Hake and Phelps[7] (Fig. 5) was based on a Born-approximation calculation and was adjusted to fit then-available swarm data. Using this cross section, we calculate values of ν_u/N which are in good agreement with all available swarm data (Fig. 6), lying within the estimated accuracy of the data. We have included this cross section in our final cross-section set[28] (Table II).

In the resonance region from 1 to 3 eV, the swarm analysis method is less sensitive to the detailed shapes of cross sections than near threshold. It is therefore necessary to rely on electron-beam-scattering measurements for the detailed shapes and relative magnitudes of the vibrational cross sections ($v=1,2,...,10$) in the resonance region. However, the method of swarm analysis is useful in determining the correct normalization of these cross sections. Ehrhardt *et al.*[25] measured relative cross sections for vibrational excitation, which they then normalized by reference to the elastic cross section. The accuracy of this normalization is difficult to estimate, since an unknown error is introduced in the necessary extrapolation to 180° of the differential cross-section data measured between 10° and 110°. Using the vibrational cross sections normalized by Ehrhardt *et al.*, our calculated values of the energy exchange collision frequency ν_u/N fall significantly below the values based on measured transport coefficients (Fig. 6). It was necessary to increase all the vibrational excitation cross sections by 90% in the energy range 1–3 eV in order to achieve the agreement with experimental swarm data shown in Fig. 6. We have adopted this normalization in our final cross-section set.

In summary, vibrational excitation of ground-state CO is described by the following cross sections: for $v=1$ (Fig. 5 and Table II), the cross section of Hake and Phelps[7,28] from threshold to 0.7 eV, smoothly joined near 1 eV to the cross section of Ehrhardt *et al.*,[25] increased by 90%. For excitation of higher levels, the cross sections of Ehrhardt *et al.* ($v=2$–7) and Boness and Schulz[29] ($v=8$–10) are increased by 90%. This cross-section set is consistent with all CO swarm data measured to date.

D. Electronic excitation

A set of cross sections for electron-impact excitation of the electronic states of CO was developed by Sawada, Sellin, and Green[30] as analytic representations based on experimental measurements. In the present analysis, those cross sections were tested for consistency with the measured electron transport coefficient data. Cross sections for excitation of the $a^3\Pi$, $A^1\Pi$, $b^3\Sigma$, $C^1\Sigma$, $E^1\Pi$, and 13.5-loss states were used (Fig. 7). The total ionization cross section used was that of Rapp and Englander-Golden.[31]

With the ionization cross section fixed, the swarm parameter most sensitive to changes in the electronic excitation cross-section set is the ionization coefficient α/N, since the electronic cross sections determine the details of the high-energy portion of the electron distribution function and, hence, the ionization rate. Using the initial set of cross sec-

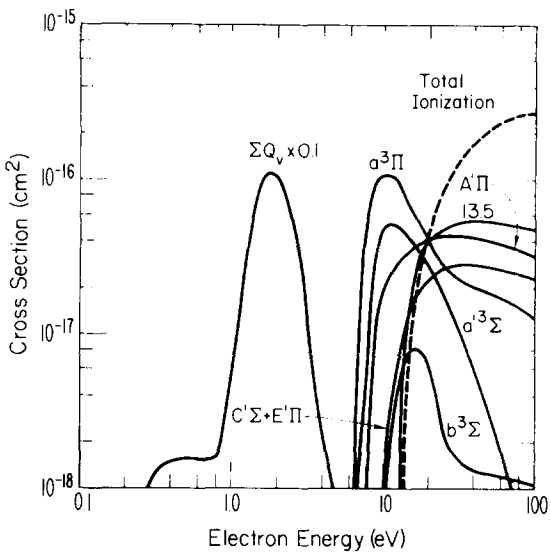

FIG. 7. Cross sections for vibrational and electronic excitation in CO determined in the present analysis. For simplicity, only the sum of the 10 vibrational cross sections is shown. The curve labeled $a'\,^3\Sigma$ is the cross section of Ref. 32 multiplied by 0.35. The remaining electronic excitation cross sections are from Ref. 30.

tions just described, the calculated values of α/N were significantly larger than measured values. This indicates that either the electronic cross sections of Sawada *et al.* are too small, or one or more electronic states are missing from the cross-section set. From a search of the literature, the only possibly missing state appeared to be one designated $a'^3\Sigma$ by Chung and Lin.[32] These authors calculated a large cross section ($\sim 1\times 10^{-16}$ cm^2 at 11 eV), although no experimental or other theoretical data are available for the state. When this cross section is included, the calculated values of α/N are much smaller than the measured values. Renormalizing the $a'^3\Sigma$ cross section downward improves the α/N fit, but it was found necessary, in addition, to introduce a negative slope into the momentum-transfer cross section at energies greater than 3 eV.[33] With this Q_m and with the $a'^3\Sigma$ cross section of Chung and Lin multiplied by 0.35, the satisfactory fit to measured α/N data shown in Fig. 3 was obtained.

Several other processes which may have an effect at large values of E/N ($>1\times 10^{-15}$ V cm^2) were examined. The inclusion of additional electronic states, such as attachment or the CI and CII states of Sawada *et al.*,[30] had no significant effect on the calculated transport coefficients. The effect on the electron energy distribution function of the low-energy secondary electrons produced by ionization was calculated using a computer program[34] which solves the time-dependent Boltzmann equation. The result was small decreases in the values of ϵ_k and α/N (4 and 7%, respectively, at $E/N=3.7\times 10^{-15}$). The convergence of the spherical harmonic expansion was tested by an approximate calculation[35] of higher terms (f_1 and f_2) in the expansion of the distribution function.[13] From these calculations, the correction[36,37] to ϵ_k is estimated to decrease the computed value of ϵ_k by 6% at $E/N=3.7\times 10^{-15}$ V cm^2. We found it necessary to apply these two corrections to the calculated data plotted in Figs. 1, 2, and 6 in order to achieve the close agreement between calculated and measured data shown there. The corrected values of ϵ_k agree with the experimental values within 5% for $E/N\leqslant 2.5\times 10^{-15}$ V cm^2 and within 10% at $E/N=3.7\times 10^{-15}$ V cm^2. We conclude that the method used in this analysis, the numerical integration of the Boltzmann equation in the spherical harmonic expansion approximation, is valid in CO for E/N as large as 4×10^{-15} V cm^2.

IV. SUMMARY

A set of cross sections for low-energy electron -CO scattering has been determined.[38] The use of collision-frequency variables isolated the effects of, and hence permitted the separate determination of, cross sections for momentum transfer and for rotational, vibrational, and electronic excitation. The set of cross sections is consistent with all available measured electron transport coefficients.

ACKNOWLEDGMENTS

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