Elastic electron collision cross sections for ammonia molecules in the energy range 0.1–1.0 keV

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Elastic differential, integral, and momentum-transfer cross sections are calculated for the e-NH_3 system in the energy range of 0.1–1.0 keV. A parameter-free spherical optical potential is constructed from near-Hartree-Fock one-center expansion ammonia wave functions. The optical potential is then treated in a partial-wave analysis to extract various cross sections. Several versions of parameter-free polarization and exchange potentials are employed. The calculated differential cross sections are compared with the available experimental data.

In this Brief Report, we present elastic differential cross sections (DCS) for the electron-NH_3 system at 0.1–1.0 keV energies, which are just extensions of our earlier calculation for the e-H_2O system (see Ref. 1). For the e-NH_3 collisions, Harshbarger *et al.*[2] measured the small-angle (2°–10°) DCS at 300, 400, and 500 eV. At very high energies, Lahmam-Bennani *et al.*[3] have measured the e-NH_3 elastic cross sections and presented them as a function of momentum transfer q. Consequently, several calculations[4–6] in the first Born approximation were published on the e-NH_3 DCS as a function of q. Jain[7] reported some preliminary calculations on the small-angle ($\theta < 10°$) DCS at 300–500 eV and obtained good agreement with the experiment of Harshbarger *et al.*[2] Experimental[8] and theoretical[9] data on the total (elastic plus inelastic) cross sections for the e-NH_3 system at intermediate and high energies have been reported very recently.

The interaction of electrons with ammonia molecules is of interest in various atomic and molecular processes occurring in plasma physics, interstellar space, atmospheric and radiation physics, etc. From the above discussion it is clear that there is a paucity of theoretical calculations on this collision system. Here our goal is to present the e-NH_3 DCS at 0.1–1.0 keV energies and compare them with experiments and other calculations. First, we briefly describe the theoretical approach.

Recently, Jain[10] has developed a simple model potential approach to calculating the total (elastic plus absorption) cross sections for electron–polyatomic-molecule systems. In this model, the total interaction of the electron-molecule system is approximated by a local spherical complex optical potential (SCOP), which is treated exactly in a partial-wave analysis to yield final cross-section quantities. The method has been very successful for those polyatomic molecules that have no dipole and quadrupole moments, e.g., the CH_4 and SiH_4 molecules (see Refs. 8 and 11). However, this restriction is meaningful at low energies only ($E < 20$ eV). At intermediate and high energies the higher-order multipole terms do not contribute significantly to the integral cross sections. Therefore an extension of the SCOP model to polar polyatomic molecules is desirable at high energies: this has recently been done by Jain[9] by employing the SCOP approximation for e-NH_3 and e-H_2O collisions at intermediate and high energies. His[9] results on the total (elastic plus inelastic) cross sections for both systems were in very good agreement with the existing experimental data. Very recently, Jain *et al.*[1] have explored the same approach using only the real optical potential for predicting the elastic DCS for the e-H_2O system. Inspired by the success of our H_2O work[1], we have repeated similar calculations for the NH_3 molecule for which experimental DCS (Refs. 2 and 3) are available.

The theoretical details are exactly the same as described earlier[1]. We would therefore not provide the full details here. In brief, assuming the adiabatic nuclei approximation,[12] the e-NH_3 effective potential $V_{opt}(\mathbf{r})$ (static plus exchange plus polarization) is calculated from molecular electronic density $\rho(r)$ and expanded around the center of mass (c.m.) of the molecule in terms of symmetry-adapted wave functions of A_1 symmetry. The first term, corresponding to $l=0$ (spherical), in the expansion of $V_{opt}(\mathbf{r})$, is treated exactly in the partial-wave decomposition scheme (we used the variable-phase approach[13] to extract the phase shifts) to yield various cross sections. The justification for neglecting the higher-order anisotropic terms (dipole, quadrupole, etc.) has already been discussed for the H_2O case[1] and the same discussion applies to the present NH_3 case also. In the present adiabatic nuclei approximation, the DCS at $\theta=0°$ are not defined. Finally, the exchange is included via the free-electron-gas exchange[14] (Hara version, to be denoted by HFEGE) and modified semiclassical exchange[15] (MSCE) models and the polarization via the correlation potential[16,17] (COP) and the energy-dependent Buckingham-type potential[18,19]. Note that there is no adjustable parameter in the present calculations.

The DCS in the energy range 0.1–1.0 keV are displayed in Figs. 1–3. Note that the cross section value at $\theta=0°$ is extrapolated. In order to see the contribution of various spherical terms in $V_{opt}(r)$, we have done calculations in several models which are abbreviated as follows: S, pure static only; SH, S plus HFEGE potential; SHP1, SH plus the correlation polarization potential; SHP2, SH plus the energy-dependent polarization poten-

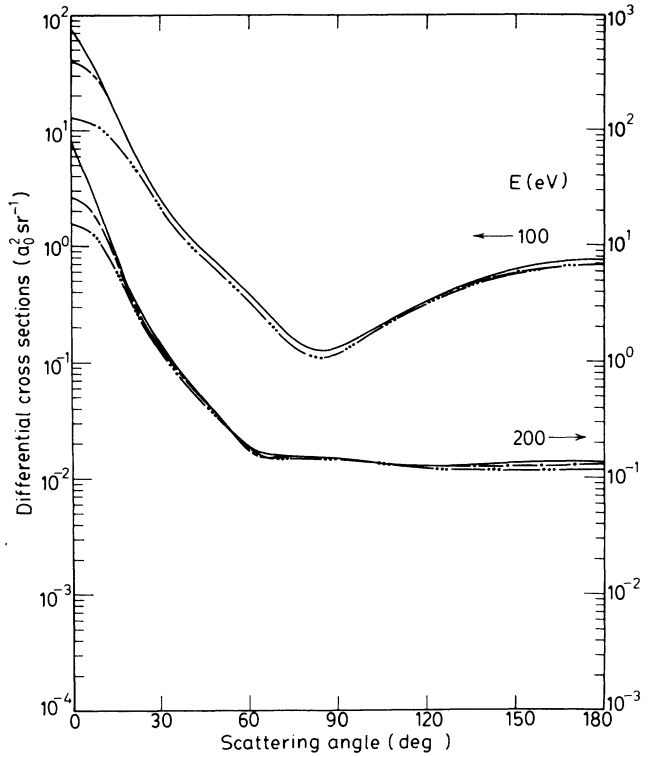

FIG. 1. Differential cross sections for the e-NH_3 scattering at 100 and 200 eV. Present calculations: ——, SHP1 model; —·—·—·, SEP1 model; —·—·, SHP2 model; —···—, S model (for notation see the text). Note the arrows for scale.

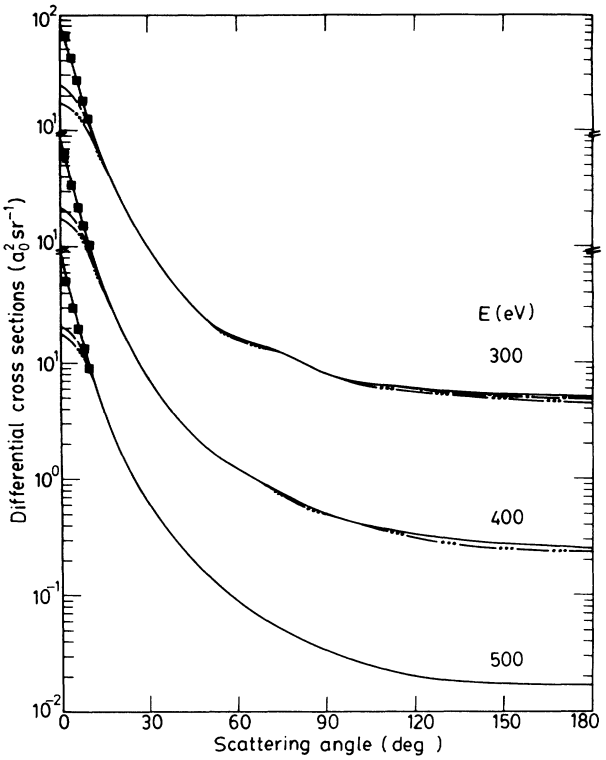

FIG. 2. Same legend as in Fig. 1 except at energies of 300, 400, and 500 eV. Experimental data: ■, Harshbarger *et al.* (Ref. 2).

tial; SE, static plus the MSCE; SEP1, SE plus the correlation polarization potential; SEP2, SE plus the energy-dependent polarization potential. The only available experimental DCS (Ref. 2) at 300, 400, and 500 eV in the forward direction (2°–10°) are also plotted in these figures. In this energy region, the SEP1 and SEP2 models are almost identical to the SHP1 and SHP2 models, respectively. This means that the two approximations for exchange (i.e., HFEGE and MSCE) are almost identical at such higher energies.

At 100 eV (Fig. 1) the shallow minimum structure at middle angles is mainly due to static interaction only. The effect of exchange and polarization together is not significant except at small angles, where polarization interaction plays an important role. Note that the present small-angle DCS (Figs. 1–3) may be slightly modified due to the long-range dipole potential which is neglected here. With the increase in energy the middle-angle minimum feature starts disappearing. There is considerable difference between the SHP1 and SHP2 models, near the forward direction. The correlation potential (SHP1 curve) is much stronger than the energy-dependent one. It is possible that the correlation polarization model is compensating for the dipole contribution: this is more clear in Fig. 2, where the present DCS at 300, 400, and 500 eV compare very well with the measurements of Harshbarger *et al.*[2] up to 10°. As noted by Jain,[7] the dipole term, when included coherently along with energy-dependent-type polarization, significantly improves the agreement with experiment[2].

At high energies, where the Born approximation is valid, the angular distribution function, plotted as a function of momentum transfer q, is independent of incident energy. Such calculations have been performed by a number of authors[4–6] in the first Born approximation employing molecular wave functions of different quality. Lahman-Bennani *et al.*[3] have measured the elastic DCS for NH_3 molecules at 35-keV incident electron energy. Figure 4 shows earlier theoretical calculations in the first Born approximation along with experimental and present results in the SHP1 model (at 1000 eV).

From Fig. 4, it is seen that the two measurements differ considerably from one another. The Harshbarger *et al.*[2] measurements are approximately 20% above at $q=0.3$ a.u. and 40% below at $q=1.0$ a.u. when compared with the measured values of Lahmam-Bennani, Duguet, and Wellenstein.[3] The reason for this discrepancy may be that the elastic DCS by Harshbarger *et al.*[2] are obtained with an incident energy of 500 eV, which is relatively smaller for the validity of the first Born approximation. It is also noticed that all the theoretical calculations differ among themselves for $q \leq 1.0$ a.u. except that the present calculation and the one-center configuration-interaction (CI) results of Tavard[6] agree well with each other in almost the entire momentum-transfer range. The present calculations are also in good accord with the measurements of Lahmam-Bennani *et al.*[3] for q values in the range 1–2 a.u., and thereafter (i.e., for $2.0 \leq q \leq 7.0$

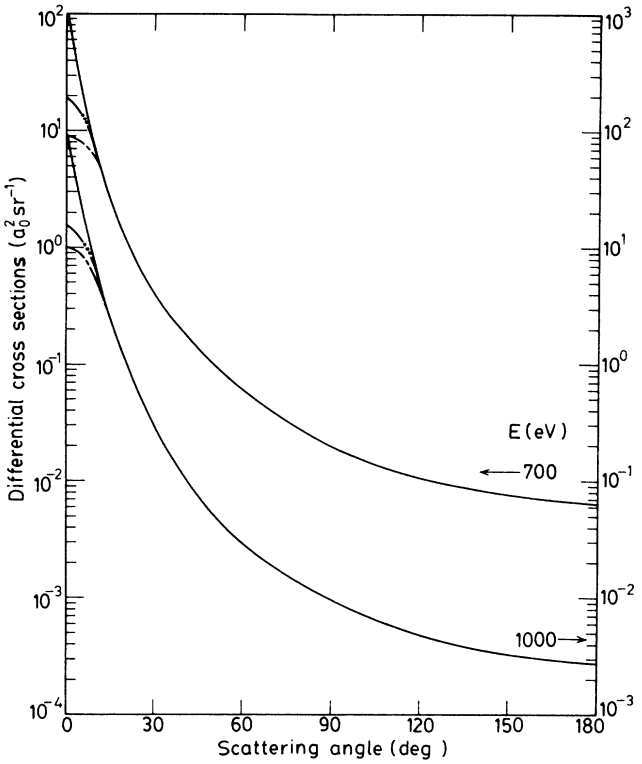

FIG. 3. Same legend as in Fig. 1 except at energies of 700 and 1000 eV (note the arrows for scale).

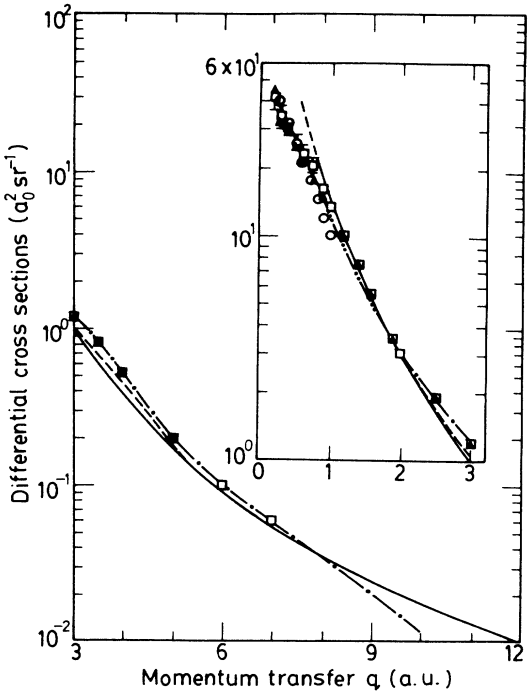

FIG. 4. Differential cross sections for the e-NH_3 elastic collisions as a function of momentum transfer q. ——, present SHP1 calculations at 1000 eV. Results in Born calculations; —··—, Szabo and Ostlund (Ref. 5); ▲, results obtained in a polarized, split valence 6-31G^* Gaussian basis set (Ref. 3); — — —, single-center configuration interaction (Ref. 6); —·—·—, Sharma and Tripathi (Ref. 4). Experimental data: □, Lahmam-Bennani *et al.* at 35 keV (Ref. 3); ○, Harshbarger *et al.* at 500 eV (Ref. 2).

a.u.) the present results underestimate the measured values approximately by 30%. In this region, the calculations of Sharma and Tripathi[4] compare reasonably well with the averaged experimental values. This clearly shows that the first Born approximation may be valid for energies well above 1000 eV; a similar conclusion was also drawn recently for the case of e-H_2O scattering[1]. Thus, for a more qualitative assessment of the high-energy limit, the absolute DCS need to be measured for the scattering of high-energy electrons.

Finally, we summarize our integral cross sections (σ_i and σ_m) which are obtained from the present DCS.

TABLE I. Elastic integral σ_i and momentum-transfer σ_m cross sections in various models for the e-NH_3 scattering in units of 10^{-16} cm^2 (for notation see the text). Values in parentheses correspond to σ_m.

Energy (eV)	Various models				
	S	SHP1	SHP2	SEP1	SEP2
100	2.65	4.15	4.02	4.16	4.03
	(1.31)	(1.44)	(1.44)	(1.40)	(1.39)
200	1.75	2.51	2.12	2.51	2.11
	(0.544)	(0.587)	(0.583)	(0.572)	(0.567)
300	1.36	1.87	1.53	1.86	1.52
	(0.308)	(0.329)	(0.327)	(0.322)	(0.320)
400	1.12	1.51	1.22	1.51	1.22
	(0.201)	(0.215)	(0.214)	(0.210)	(0.209)
500	0.954	1.28	1.03	1.27	1.03
	(0.144)	(0.153)	(0.152)	(0.150)	(0.149)
700	0.743	0.977	0.731	0.945	0.729
	(0.0847)	(0.0895)	(0.0966)	(0.0879)	(0.0951)
1000	0.553	0.747	0.551	0.746	0.549
	(0.0488)	(0.0506)	(0.0535)	(0.0498)	(0.0528)

(Thus our σ_i values are finite in the present adiabatic approximation.) There exist no experimental results to compare with our σ_i and σ_m values. We have calculated the σ_i and σ_m cross sections in our various models and the same are tabulated in Table I. From this table it is seen that the σ_i and σ_m cross sections are quite sensitive to the choice of different polarization potential but not to the exchange approximations.

In conclusion, we have reported differential (and integrated total and momentum-transfer) cross sections for the e-NH_3 elastic scattering at 0.1–1.0 keV energies. A parameter-free spherical optical potential for the e-NH_3 system is treated exactly in a partial-wave scheme to determine the final cross sections. The optical potential includes exchange and polarization effects. These results compare very well with experimental data wherever they are available. The higher-order anisotropic potential terms (dipole, quadrupole, etc.) are neglected here with the assumption that such effects are negligible at such high energies and scattering angles except near the forward direction ($\theta \geq 5°$).

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