

Vibrationally elastic scattering of positrons from CH₄ molecules

Tamio Nishimura*
Atomic Physics Laboratory

Introduction

Here we report our theoretical study on the vibrationally elastic scattering of positrons (e^+) from methane (CH₄) molecules. This is an extension of our previous work on $e^- + \text{CH}_4$ collision.¹⁾

Making use of a positron as a projectile instead of an electron supplies some interesting phenomena in the field of atomic physics. For instance, the incident positron is distinguishable from molecular electrons. In other words, there is no effect of electron exchange considered in the case of electron-molecule collision. In addition, the positron has a possibility of taking an electron from the target molecule and making an positronium (Ps) with many kinds of quantum states, provided that the collision energy is beyond the Ps formation threshold (5.8 eV for CH₄). From a theoretical point of view, the DCS, in particular for the positron scattering, has been found to depend sensitively on the target polarization effects.²⁾

On the other hand, total cross sections (TCSs) on the $e^+ + \text{CH}_4$ system have ever been measured several times.^{3,4)} Besides, differential cross sections (DCSs) on quasielastic process (i.e., purely elastic scattering plus rotational and vibrational excitations) for the system also have been measured very recently for the first time in the collision energies of 4-200 eV by the group of Kauppila.⁵⁾ The appearance of these experimental cross sections and the theoretical interests mentioned above strongly motivate the author to test the validity of the present theory^{1,6)} for the case of positron. Here, atomic units (au) are used unless otherwise stated.

Theory

The details of the present theory have been already reported in our previous paper.⁹⁾ The present theory is based on the fixed-nuclei approximation. In order to properly consider the molecular symmetry (Td point group for CH₄), the wavefunction (F) of scattered positron is expanded in terms of the symmetry-adapted angular basis functions (X_l^ν),⁷⁾ as follows:

$$F(\mathbf{r}) = \frac{1}{r} \sum_{l\nu} f_{l\nu}(r) X_l^\nu(\hat{\mathbf{r}}), \quad (1)$$

where \mathbf{r} denotes the position vector of incident positron. The index ν stands for indices collectively specifying an irreducible representation (IR) and those distinguishing the degenerated members of the same l and IR.

The interaction potential (V) between the positron and the CH₄ molecule is represented in the form of a local potential which consists of the repulsive electrostatic (V^{st}) and the attractive correlation-polarization (V^{pol}) potentials. Since both of these potentials have opposite signs each other for the positron scattering, cancellation effects are caused and lead to the collision system very sensitive to V^{pol} potential. As for V^{pol} , the model potential (V^{corr}) which is based on the correlation energy of a positron in a homogeneous electron gas⁸⁾ is adopted in the short-range region of r . The V^{corr} is smoothly connected with the asymptotic form of $-\alpha_0/2r^4$ with α_0 being the polarizability of CH₄. The present potentials (V^{st} and V^{pol}) both depend only on the undistorted electron density of the molecule. In a manner similar to Eq. (1), the interaction matrix is expanded as

$$\langle l\nu | V | l'\nu' \rangle = \sum_{\lambda\mu} V_{\lambda\mu}(r) \times \int d\hat{\mathbf{r}} X_l^{\nu*}(\hat{\mathbf{r}}) X_\lambda^\mu(\hat{\mathbf{r}}) X_{l'}^{\nu'}(\hat{\mathbf{r}}). \quad (2)$$

Then, the radial part $f(r)$ in Eq. (1) satisfies a set of coupled differential equations:

$$\left\{ \frac{d^2}{dr^2} - \frac{l(l+1)}{r^2} + k^2 \right\} f_{l\nu}(r) = 2 \sum_{l'\nu'} \langle l\nu | V | l'\nu' \rangle f_{l'\nu'}(r). \quad (3)$$

By solving Eq. (3) with an appropriate boundary condition, we obtain the scattering matrix, from which the cross sections are determined.

Numerical calculations

In the present calculation, the molecular orbital code GAMESS⁹⁾ is employed to generate a multi-centered target wavefunction. The target wavefunction for the CH₄ molecule is exactly same as that used in the case of $e^- + \text{CH}_4$.¹⁾ For the asymptotic part of V^{pol} , the experimental value of the polarizability ($\alpha_0 = 17.97$ au) is adopted. In the expansion of Eq. (2), we retained the terms up to $\lambda = 8$. The coupled equations (3) are solved with inclusion of partial waves up to $l = 8$ for each of the IRs in the Td point group. For partial waves higher than this, the polarized first-Born approximation is used to obtain the relevant elements of the T -matrix. The DCS was calculated in the region of the collision energies from 1 to 10 eV. In the present calculation, the Ps formation channel which is open in the collision energy larger than 5.8 eV is neglected.

* tamimisi@postman.riken.go.jp

Results and discussion

1. Integral cross section Figure 1 shows integral cross sections (ICSs) as a function of the collision energy. The present vibrationally elastic ICS is shown together with the present one obtained with the electrostatic potential V^{st} only to see the contribution of polarization effect. From Fig. 1, the effect of the V^{pol} is of great significance all over the collision energies calculated. The present ICS is compared with the experimental TCS by Sueoka and Mori.³⁾ The values of present ICS are too large in comparison with those of the experimental TCS. In principle, vibrationally elastic ICS must be less than TCS in magnitude at a constant collision energy, because the latter quantity also includes all of the contribution from inelastic processes. These inconsistencies may be attributed to insufficient accuracy of the local model potential V^{corr} of the V^{pol} introduced.

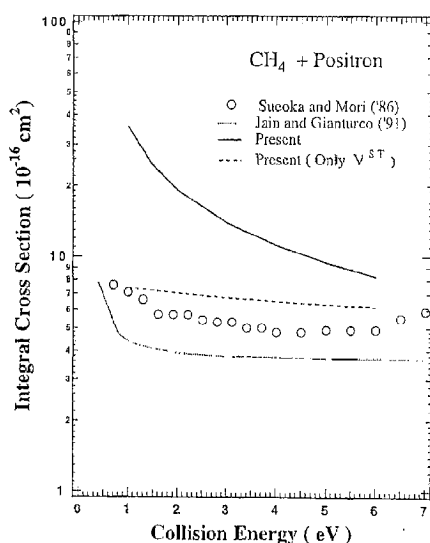


Fig. 1. Integral cross section (ICS) (in 10^{-16}cm^2) on the $e^+ + \text{CH}_4$ system as a function of the collision energy (in eV). The present (vibrationally elastic) ICS with the electrostatic and the polarization potentials is compared with the present one with the electrostatic potential only, the other calculation by Jain and Gianturco and the total cross section measured by Sueoka and Mori.

Moreover, a comparison is also made with the other theoretical cross section by Jain and Gianturco.²⁾ The procedure of present calculation is almost same as Jain and Gianturco except for minor technical points, however again the results of the both calculations disagree considerably with each other. They employ a single-centered target wavefunction,¹⁰⁾ which will give rise to a rather different electron density near the target nuclei from the multi-centered wavefunction used in the present calculation. The difference between the two kinds of wavefunctions is probably amplified especially in the V^{corr} , which leads to a great discrepancy of the final results between the two. Maybe, a better way of taking the polarization effect into account than the present one and an appropriate inclusion of Ps formation channel into the close-coupling equations will further improve the present cross sections quantitatively.

2. Differential cross section Figure 2 shows the DCSs at the collision energies of 4, 6 and 10 eV. The resulting DCSs are compared with those measured by Kauppila and co-workers.⁵⁾

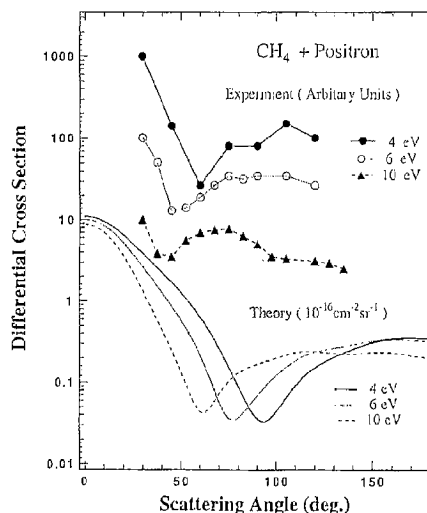


Fig. 2. Differential cross sections (DCSs) on the $e^+ + \text{CH}_4$ system at the collision energies of 4, 6 and 10 eV. The present (vibrationally elastic) DCSs shown by curves are respectively compared with the (quasi elastic) experimental ones by symbols.

Their DCSs are in a relative scale and they claim that their data correspond to quasielastic scattering process which is equivalent to electronically elastic one. In the experimental DCS, a notable minimum is observed at around 60° at 4 eV, and its position moves to be at the smaller angle as the energy increases. We compare the shape of the experimental DCSs for so-called *electronically* elastic process with the present ones for the *vibrationally* elastic one, because generally a contribution of vibrational excitation cross section has been found to be small to vibrationally elastic one.

It should be noted that throughout the collision energy calculated, the shapes of the experimental DCSs are roughly reproduced by the present calculation, although the positions of the minima given by present calculation are rather different from those observed in the experimental data. In addition, we find out that the shape of DCS (including the position of the minimum) is almost determined by a superposition of $l = 1$ and $l = 2$ partial waves. A better accordance of theoretical DCS in shape with the experimental one will be seen by means of an improvement of V^{pol} potential and an introduction of the Ps formation channel which is neglected in the present calculation.

References

- 1) T. Nishimura and Y. Itikawa: J. Phys. B; At. Mol. Opt. Phys. **27**, 2309 (1994).
- 2) A. Jain and F. A. Gianturco: J. Phys. B; At. Mol. Opt. Phys. **24**, 2387 (1991).
- 3) O. Sueoka and S. Mori: J. Phys. B; At. Mol. Phys. **19**, 4035 (1986).
- 4) M. S. Dababneh et al.: Phys. Rev. A **38**, 1207 (1988).
- 5) D. A. Przybyla et al.: Phys. Rev. A **55**, 4244 (1997).
- 6) Y. Okamoto et al.: J. Phys. B; At. Mol. Opt. Phys. **26**, 745 (1993).
- 7) P. G. Burke et al.: J. Phys. B; At. Mol. Phys. **5**, 2212 (1972).
- 8) E. Boronski and R. M. Nieminen: Phys. Rev. B **34**, 3820 (1986).
- 9) M. W. Schmidt et al.: J. Comp. Chem. **14**, 1347 (1993).
- 10) F. A. Gianturco and D. G. Thompson: Chem. Phys. **14**, 111 (1976).