Electronic excitation to low-lying states of GeF$_4$ molecule by electron impact: A comparative study with CF$_4$ and SiF$_4$ molecules

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Electronic excitation to low-lying states of GeF₄ molecule by electron impact: 
A comparative study with CF₄ and SiF₄ molecules

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Synopsis We report on the measurements of the electron impact electronic excitation cross sections for XF₄ (X = C, Si and Ge) molecules at 100 eV, 5° scattering angle and 30 eV, 30° in the electron energy loss range 8.0 – 18 eV. For a target of GeF₄ molecule, the optically-forbidden behavior has been observed in the lower electron energy loss range.

CF₄ is one of the more commonly used etchant gases for dry-etching process, where the fluorine atoms created in CF₄/O₂ mixing plasma discharged on silicon surfaces, form SiF₄ molecule. GeF₄ molecule has been also used for the manufacturing of semiconductors. In spectroscopic studies, electron energy loss spectra (EELS) and photoabsorption cross sections of XF₄ (X = C, Si and Ge) molecules were obtained for understanding the optical diagnostics of chemical process in etching plasmas, e.g. refs [1, 2] and references therein. Recently, an angle-resolved EELS study in CF₄ has been reported [3]. Our group have also carried out a series of experimental measurements to probed the scattering dynamics of low energy electron collisions with tetrahedral (Tₐ) symmetry molecules such as CF₄ [4], SiF₄ [5] and GeF₄ [6]. In this work, we report on the measurements of the electron impact excitation cross sections for XF₄ molecules at 100 eV, 5° scattering angle and 30 eV, 30° by electron energy loss spectroscopy in order to observe the contribution of the optically-forbidden transitions.

The electron energy loss spectrometer used in this work has been described sufficiently in previous publication [7]. In brief, the spectrometer is a crossed-beam type with a hemispherical monochromator and analyzer pumped differentially and electron lens system controlled by computer-driven voltages. The setup was operated at impact energies from 1.5 to 300 eV and a scattering angle range from -6° to 130°, with typical energy resolution of 40 – 45 meV (FWHM). The electron energy scale was calibrated against the 19.37 eV Feshbach resonance in He atom and the angular scale were determined from the symmetry in the intensity profile of the He 2¹P excitation measured at the 0° nominal scattering angle. The absolute scale of the excitation cross sections has been obtained by the relative flow technique with He elastic differential cross sections as the reference species.

Comparison between electron energy loss spectra of XF₄ molecules at 100 eV incident energy, 5° scattering angle and at 30 eV, 30° have shown clear differences. In the case of the GeF₄ molecule, a shoulder structure is discernible and due to the optically-forbidden transition in the lower electron energy loss range than the lowest-lying optically-allowed transition, which have already assigned as 4e⁺ transition.

Detailed discussion of the comparison among the electron energy loss spectra of XF₄ molecules and quantum chemical calculations, basically, EOM-CCSD method with aug-cc-pVQZ + Rydberg basis set using MOLPRO code will be presented at the conference.

References

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