Supporting information

Supporting Information for the paper entitled "A Strong Charge-Transfer Effect in Surface-Enhanced Raman Scattering Induced by Valence Electrons of Actinide Elements" by Yang Gao, Lei Chen, Xing Dai, Ruixia Song, Bo Wang and Zhigang Wang

Contents

Part 1. The absorption spectra of six complexes.

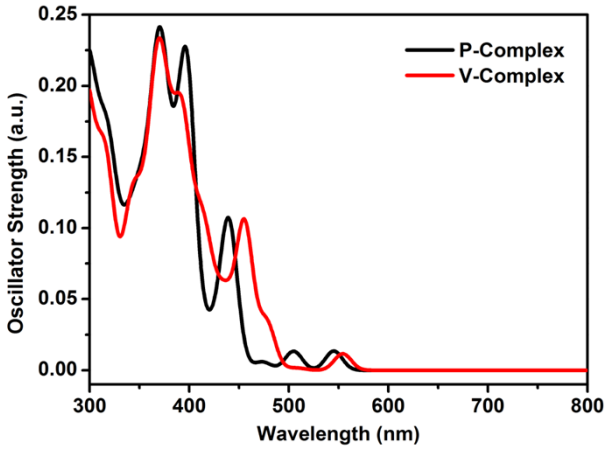

Figure S1. The absorption spectra of P-complex and V-complex. Absorption coefficients are given as the oscillator strength in a.u. and wavelength in nm. Spectra have been broadened by a Lorentzian having a width of 20 cm^{-1}.

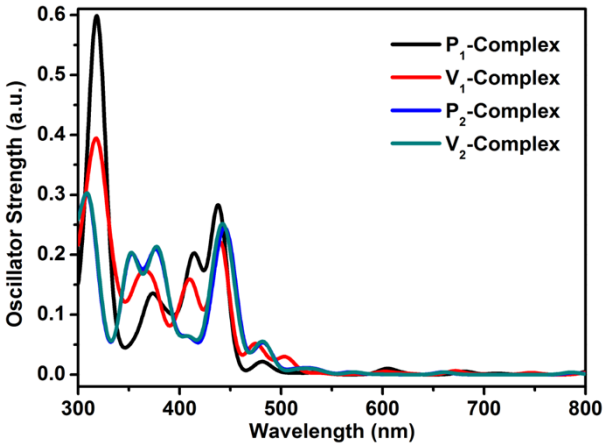

Figure S2. The absorption spectra of P_1-complex, V_1-complex, P_2-complex and V_2-complex.

For the absorption spectra of the six complexes, the peaks of oscillator strength are very weak in the long excitation wavelength region (see Figure S1 and Figure S2). Especially for P-complex and V-complex, which have no even the presence of transition (f = 0.0) when the wavelength is greater than 550 nm (see Figure S1). Therefore, in this paper, we don't discussed the long excitation wavelength region.

Part 2. the additional CT excitation energies of six complexes.

Table S1. Calculated excitation energies (E in eV), excitation wavelengths (λ in nm), oscillator strengths (f) and orbital transitions of CT excitations for six complexes, respectively.

Geometry	E / λ	f	Transitions
P-complex	2.48 / 499	0.0002	HOMO → LUMO+2
V-complex	3.02 / 411	0.0607	HOMO-2 → LUMO+2
P_1-complex	2.77 / 447	0.0014	HOMO-5 → LUMO+2
V_1-complex	3.41 / 363	0.0166	HOMO-20 → LUMO+2
P_2-complex	3.24 / 382	0.0455	HOMO-16 → LUMO+2
V_2-complex	325 / 381	0.0688	HOMO-16 → LUMO+2

Part 3. The IEs of pyridine-Ac@Au_7 and pyridine-Au_8 complexes.

Table S2. Calculated excitation energies (E in eV), excitation wavelengths (λ in nm), oscillator strengths (f) and orbital transitions of IEs for six complexes, respectively.

Geometry	E / λ	f	Transitions
P-complex	3.36 / 369	0.1689	HOMO → LUMO+7
V-complex	2.60 / 478	0.0331	HOMO → LUMO+3
P_1-complex	3.08 / 403	0.0270	HOMO-13 → LUMO+1
V_1-complex	2.99 / 414	0.0348	HOMO-10 → LUMO+1
P_2-complex	2.82 / 439	0.1006	HOMO-16 → LUMO
V_2-complex	2.75 / 450	0.0941	HOMO-16 → LUMO

Part 4. Calculated static Raman spectra of pyridine-Au_8 complexes.

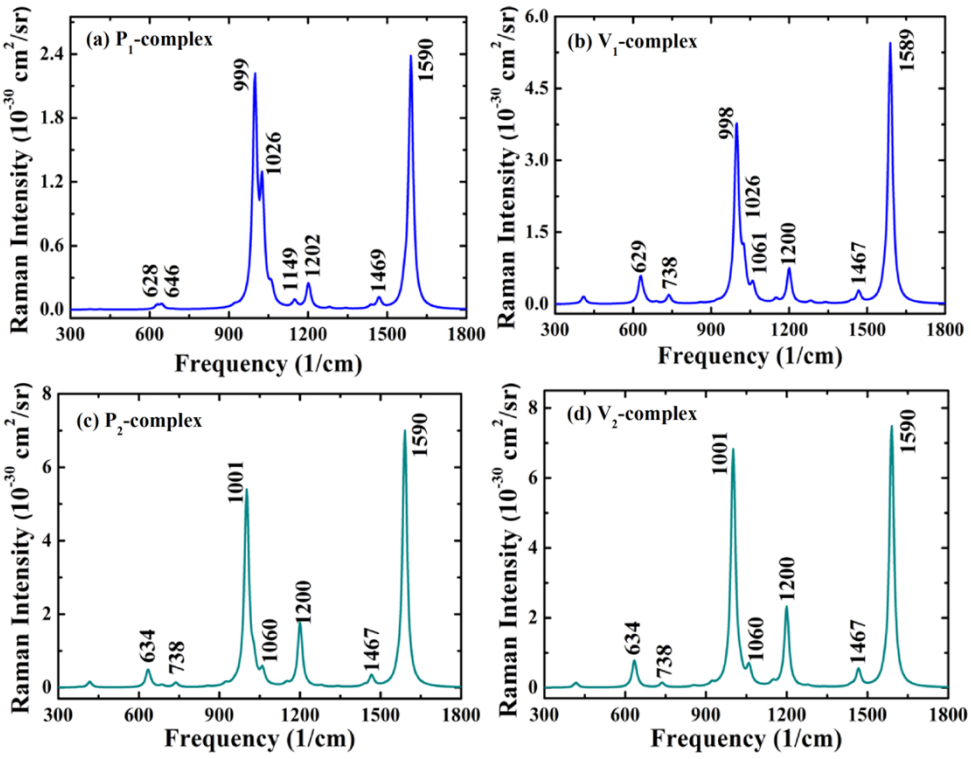

Fig S3. Calculated static Raman spectra of (a) P_1-complex, (b) V_1-complex, (c) P_2-complex, (d) V_2-complex. Differential cross-section is measured in the unit of 10^{-30} cm^2/sr. Spectra have been broadened by a Lorentzian having a width of 20 cm^{-1}.