A ^{35}Cl NQR Study on $Cs_2[Au^ICl_2][Au^{III}Cl_4]$*

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A pair of ^{35}Cl NQR spin echo signals has been observed for the mixed valence complex $Cs_2[Au^ICl_2][Au^{III}Cl_4]$ between 77 and 243 K. At 77 K, two resonance lines with the half widths $\Delta\nu_Q \sim 50$ kHz were located at $\nu_{Q1} = 17.28$ MHz for the Au^I-Cl chlorine and at $\nu_{Q2} = 27.10$ MHz for the Au^{III}-Cl chlorine in accordance with the crystal structure. The chlorine ionic characters of the Au^I-Cl and Au^{III}-Cl bonds are estimated as 0.63 and 0.42, respectively. The central gold atom carries a fractional protonic charge of 0.26 in $[Au^ICl_2]^-$ and 0.68 in $[Au^{III}Cl_4]^-$. The charge distributions in the complex anions differ insignificantly from those in the isolated $[AuCl_2]^-$ and $[AuCl_4]^-$ for ordinary complexes, indicating that the charge transfer interactions between the anions are weak in the mixed valence complex. The observed linear temperature dependencies of ν_Q and $\log T_{1Q}$ are well explained by the lattice vibration. When the temperature was increased from 77 K, the resonance lines became gradually weak without changing $\Delta\nu_Q$ and immeasurable above 215 K. ESR spectra taken at various temperatures revealed the presence of paramagnetic sites of ca. 5×10^{20} mol^{-1} arising from Au(II). The small but finite concentration of Au(II) or some other reason should be responsible for the fade out phenomenon and the large $\Delta\nu_Q$ observed.

Key words: ^{35}Cl NQR Frequency, ^{35}Cl NQR Spin-lattice Relaxation, Mixed Valence Gold Complex, Charge Transfer Interaction, ESR.

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