The red dye Sudan I was investigated by Raman spectroscopy using different excitation wavelengths (1064, 532 and 244 nm). A calculation of the Raman spectrum based on quantum mechanical ab initio density functional theory (DFT) was made using the RB3LYP method with the 3-21G and 6-311+G(d,p) basis sets. The vibrations in the region 1600–1000 cm\(^{-1}\) were found to comprise various mixed modes including in-plane stretching and bending of various C–C, N–N, C–N and C–O bonds and angles in the molecule. Below ∼900 cm\(^{-1}\), the out-of-plane bending modes were dominant. The central hydrazo chromophore of the Sudan I molecule was involved in the majority of the vibrations through N–N and C–N stretching and various bending modes. Low-intensity bands in the lower wavenumber range (at about 721, 616, 463 and 218 cm\(^{-1}\)) were selectively enhanced by the resonance Raman effect when using the 532 nm excitation line. Comparison was made with other azo dyes in the literature on natural, abundant plant pigments. The results show that there is a possibility in foodstuff analysis to distinguish Sudan I from other dyes by using Raman spectroscopy with more than one laser wavelength for resonance enhancement of the different bands.