We develop an algorithm to obtain the natural spin-orbitals (natural spinors) for the \( L-S \) coupled wave functions. These natural spinors are generally complex-valued, mixing two spin components, and they have similar symmetry properties as the spinors from the \( j-j \) coupling calculations. Therefore, they can serve as an approximation to the spinors and any wave function analysis based on the \( j-j \) spinors can be carried out using the natural spinors. The comparison between the natural spinors and the \( j-j \) spinors of three representative atoms, Tl, At, and Lu shows their close similarity. The anti-bonding spin-orbit effects on TlH and Tl\(_2\) are studied using these natural spinors and it is found that the spin-orbit induced electron rotation from the bonding to the non-bonding or anti-bonding orbitals has a large effect on the bond strength. The natural spinors are also used to explain the spin-orbit quenching of the Jahn-Teller effect. By taking WF\(_5\) as an example (see the attached figure), we show that the spin-orbit interaction can lead to stabilization of the totally symmetric electron distribution thus stabilizing the higher symmetry molecular structure. Using the example of WF\(_6\) anion we apply the natural spinor technique to explain the Jahn-Teller distortion on the adiabatic energy surfaces that include spin-orbit coupling.

Figure 1: Structures and natural orbitals of WF\(_5\): (a) Structure of WF\(_5\); (b) \( e''_{xz} \) orbital; (c) \( e''_{yz} \) orbital; (d) Top-view of the spin-orbit electron transfer between the two \( e'' \) orbitals; (e) Electron density from the \( e_{1/2,1/2} \) natural spinor (other spinors have the same density due to symmetry). The \( \hat{l}_z \) is centred at W.