The propensity of the NO radical to dimerize has long been known. From pure rotational\textsuperscript{a} and infrared\textsuperscript{b} spectra, we know that (NO\textsubscript{2}) is a planar $C\textsubscript{2v}$ molecule with an N-N distance of about 2.26 Å and a singlet ground electronic state. One mystery of the NO dimer involves the nature and location of the many other dimer electronic states (both singlet and triplet) that are formed by bringing together two NO monomers. Another mystery involves the locations of the four low frequency (intermolecular) vibrational modes of the dimer, which have never previously been observed in the gas phase. We have now solved the second mystery by means of observations with a long-path (200 m), low-temperature (90 K) absorption cell and a Bomem FTIR spectrometer in both the mid- and far-infrared regions. The $A\textsubscript{1}$ vibrations $v\textsubscript{2}$ (symmetric bend) and $v\textsubscript{3}$ (intermolecular stretch) have been unambiguously assigned at 239.36 and 134.50 cm\textsuperscript{-1}, respectively. The very weak $v\textsubscript{3}$ ($b\textsubscript{2}$ antisymmetric bend) band is located at 429.14 cm\textsuperscript{-1}. And the infrared-forbidden $v\textsubscript{4}$ ($a\textsubscript{2}$ torsion) vibration is at about 117 cm\textsuperscript{-1}. These values for the intermolecular modes of (NO\textsubscript{2}) agree poorly with previous “accepted” values\textsuperscript{c} based on spectra of condensed NO or matrix-isolated NO dimer. However, they are consistent with a force field which we had derived\textsuperscript{d} from centrifugal distortion parameters. The NO dimer poses a challenge for quantum chemists, as will be shown by comparing our results with some recent calculations.


