

Supplementary Material Fig. 35: (a) Plot of rate coefficients  $k_{Ar}^{\Delta J}$  for rotationally inelastic collisions of NaCs 2(*A*)<sup>1</sup> $\Sigma^+$ (*v*=14, *J*=32) molecules with argon atoms, comparing the results of fits obtained with various fixed values of  $k_{Cs}^{Q}/\Gamma$  or with  $k_{Cs}^{Q}/\Gamma$  allowed to vary. (b) Plot of rate coefficients for rotationally inelastic collisions of NaCs 2(*A*)<sup>1</sup> $\Sigma^+$ (*v*=14, *J*=32) molecules with cesium atoms, in units of the cesium quenching rate coefficient  $k_{Cs}^{Q}/\Gamma$  comparing the results of fits obtained with various fixed values of  $k_{Cs}^{Q}/\Gamma$  or with  $k_{Cs}^{Q}/\Gamma$  allowed to vary.  $k_{Cs}^{Q}/\Gamma$  values are in units of cm<sup>3</sup>. For cases listed as "vary  $k_{Cs}^{Q}/\Gamma$  within limits" the range of allowed values was  $1 \times 10^{-17}$  cm<sup>3</sup>  $< k_{Cs}^{Q}/\Gamma < 1 \times 10^{-15}$  cm<sup>3</sup>. The value of  $\Gamma$  was taken to be 2.82  $\times 10^7$  s<sup>-1</sup>.





Supplementary Material Fig. 36: Plots of fluorescence ratio data ( $R_F$ ) versus argon density for  $\Delta J = +1$  collisions of NaCs 2(A)<sup>1</sup> $\Sigma^+$ (v=14, J=32) molecules with argon and cesium perturbers. Each panel represents a fixed cesium density  $n_{Cs}$ .

 $R_F$  vs.  $n_{Ar}$  for NaCs 2(A)<sup>1</sup> $\Sigma^+$ (v=14, J=32),  $\Delta J$  = +2



Supplementary Material Fig. 37: Plots of fluorescence ratio data ( $R_F$ ) versus argon density for  $\Delta J = +2$  collisions of NaCs 2(A)<sup>1</sup> $\Sigma$ <sup>+</sup>(v=14, J=32) molecules with argon and cesium perturbers. Each panel represents a fixed cesium density  $n_{Cs}$ .

 $R_F$  vs.  $n_{Ar}$  for NaCs 2(A)<sup>1</sup> $\Sigma^+$ (v=14, J=32),  $\Delta J$  = +3



Supplementary Material Fig. 38: Plots of fluorescence ratio data ( $R_F$ ) versus argon density for  $\Delta J = +3$  collisions of NaCs 2(A)<sup>1</sup> $\Sigma^+$ (v=14, J=32) molecules with argon and cesium perturbers. Each panel represents a fixed cesium density  $n_{Cs}$ .

 $R_F$  vs.  $n_{Ar}$  for NaCs 2(A)<sup>1</sup> $\Sigma^+$ (v=14, J=32),  $\Delta J$  = +4



Supplementary Material Fig. 39: Plots of fluorescence ratio data ( $R_F$ ) versus argon density for  $\Delta J = +4$  collisions of NaCs 2(A)<sup>1</sup> $\Sigma$ <sup>+</sup>(v=14, J=32) molecules with argon and cesium perturbers. Each panel represents a fixed cesium density  $n_{Cs}$ .





Supplementary Material Fig. 40: Plots of fluorescence ratio data ( $R_F$ ) versus argon density for  $\Delta J = -1$  collisions of NaCs 2(A)<sup>1</sup> $\Sigma^+$ (v=14, J=32) molecules with argon and cesium perturbers. Each panel represents a fixed cesium density  $n_{Cs}$ .

 $R_F$  vs.  $n_{Ar}$  for NaCs 2(A)<sup>1</sup> $\Sigma^+$ (v=14, J=32),  $\Delta J = -2$ 



Supplementary Material Fig. 41: Plots of fluorescence ratio data ( $R_F$ ) versus argon density for  $\Delta J = -2$  collisions of NaCs 2(A)<sup>1</sup> $\Sigma$ <sup>+</sup>(v=14, J=32) molecules with argon and cesium perturbers. Each panel represents a fixed cesium density  $n_{Cs}$ .



Supplementary Material Fig. 42: Plots of fluorescence ratio data ( $R_F$ ) versus argon density for  $\Delta J = -3$  collisions of NaCs 2(A)<sup>1</sup> $\Sigma^+$ (v=14, J=32) molecules with argon and cesium perturbers. Each panel represents a fixed cesium density  $n_{Cs}$ .

 $R_F$  vs.  $n_{Ar}$  for NaCs 2(A)<sup>1</sup> $\Sigma^+$ (v=14, J=32),  $\Delta J = -4$ 



Supplementary Material Fig. 43: Plots of fluorescence ratio data ( $R_F$ ) versus argon density for  $\Delta J = -4$  collisions of NaCs 2(A)<sup>1</sup> $\Sigma$ <sup>+</sup>(v=14, J=32) molecules with argon and cesium perturbers. Each panel represents a fixed cesium density  $n_{Cs}$ .



Supplementary Materials Fig. 44: Plot of NaCs excitation spectra for two different cesium densities and similar argon densities  $(n_{Ar} \sim 6.0 \times 10^{16} \text{ cm}^{-3})$  comparing the difference in relative peak intensities for  $\Delta J = +1$  and  $\Delta J = -1$  collisional lines. a)  $n_{Cs} = 1.04 \times 10^{16} \text{ cm}^{-3}$ ,  $I_{col}^{\Delta J=+1}/I_{col}^{\Delta J=-1} = 0.78$ . b)  $n_{Cs} = 4.37 \times 10^{16} \text{ cm}^{-3}$ ,  $I_{col}^{\Delta J=+1}/I_{col}^{\Delta J=-1} = 0.62$ .



Supplementary Materials Fig. 45: Spectra showing  $\Delta J = \pm 1, \pm 2$  NaCs  $2(A)^1 \Sigma^+(v = 14, J = 32)$  rotationally inelastic collisions with argon and helium buffer gases. The top trace shows a spectrum obtained using argon as the buffer gas, where a decrease in the intensity of the collisional peaks with increasing  $|\Delta J|$  can be seen. The bottom trace shows the same NaCs collisional transitions, except with helium as the buffer gas. The number of  $\Delta J = \pm 2$  collisions, relative to the number of  $\Delta J = \pm 1$  collisions, appears to be greater for helium than for argon perturbers.