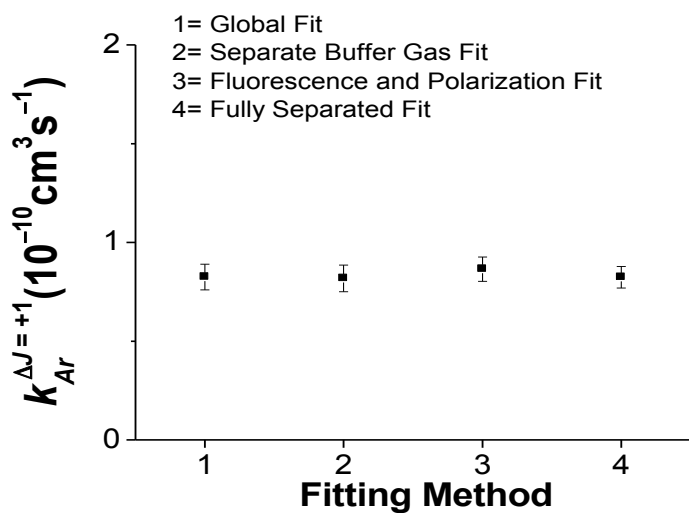
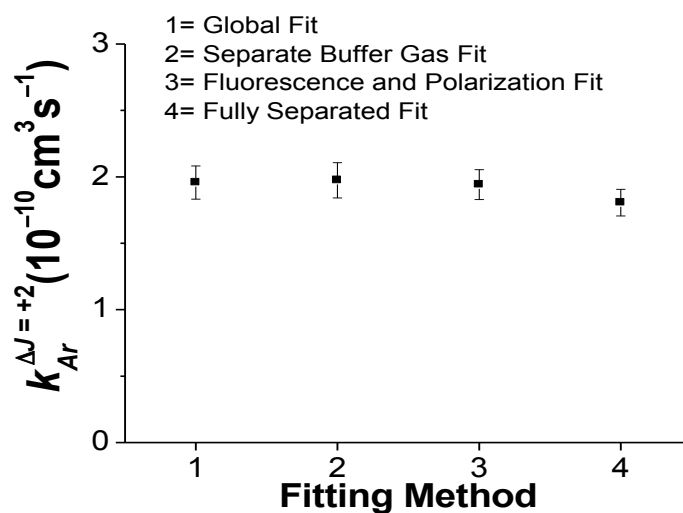


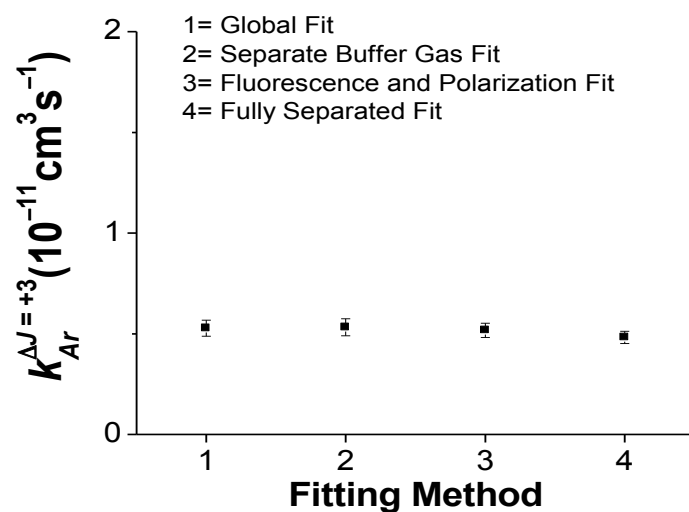
# $k_{Ar}^{\Delta J}$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$



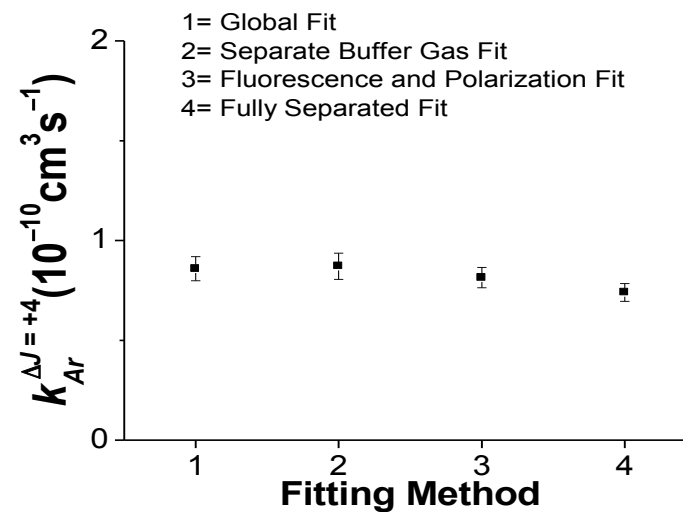
(a)  $\Delta J = +1$



(b)  $\Delta J = +2$



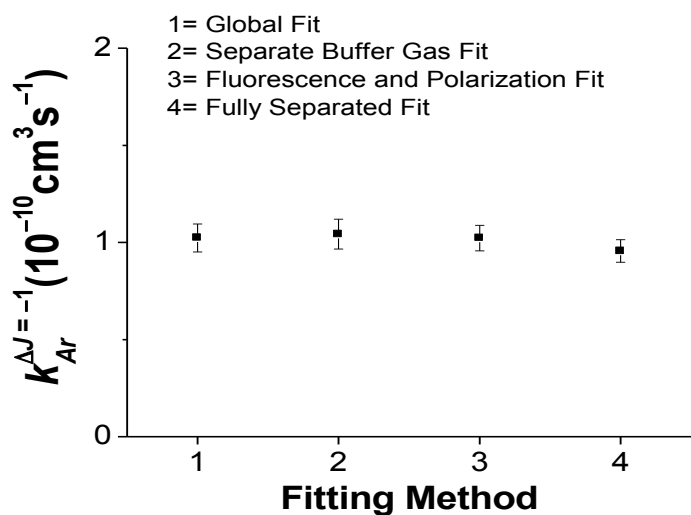
(c)  $\Delta J = +3$



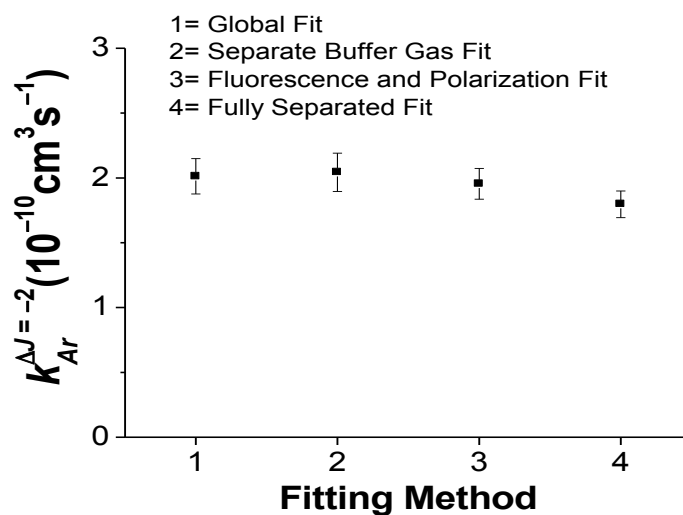
(d)  $\Delta J = +4$

Supplementary Material Fig. 1a: Plots of the fitted argon  $J$ -changing rate coefficients  $k_{Ar}^{\Delta J}$  following excitation of NaK molecules to the  $2(A)^1\Sigma^+(v=16, J=30)$  level, for each of the fitting methods described in the text ( $+4 \geq \Delta J \geq +1$ ).

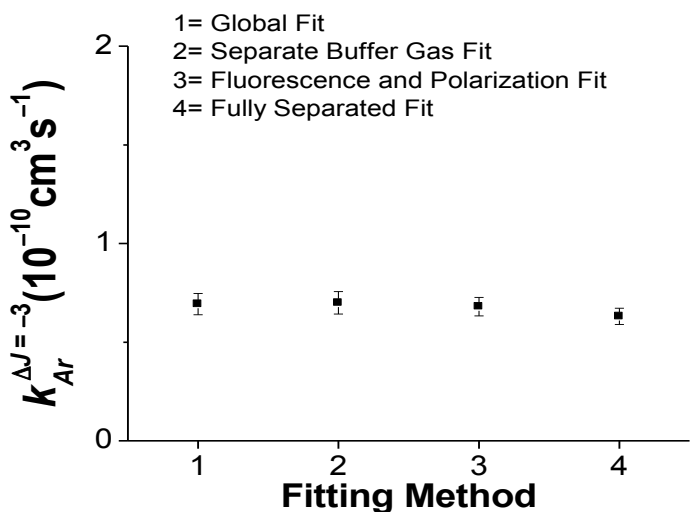
# $k_{Ar}^{\Delta J}$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$



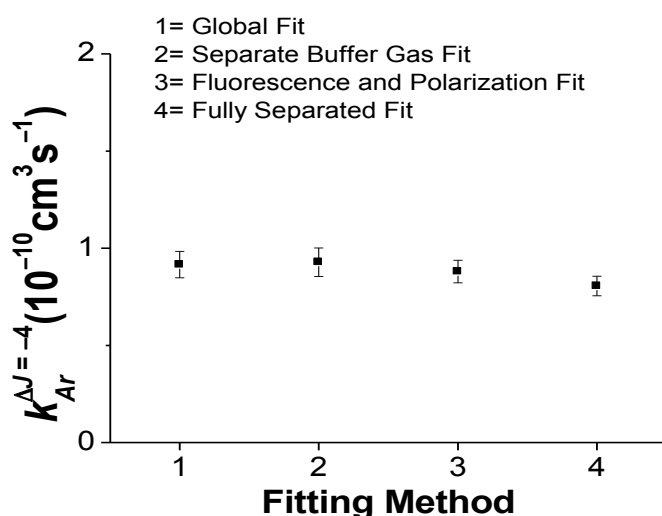
(a)  $\Delta J = -1$



(b)  $\Delta J = -2$



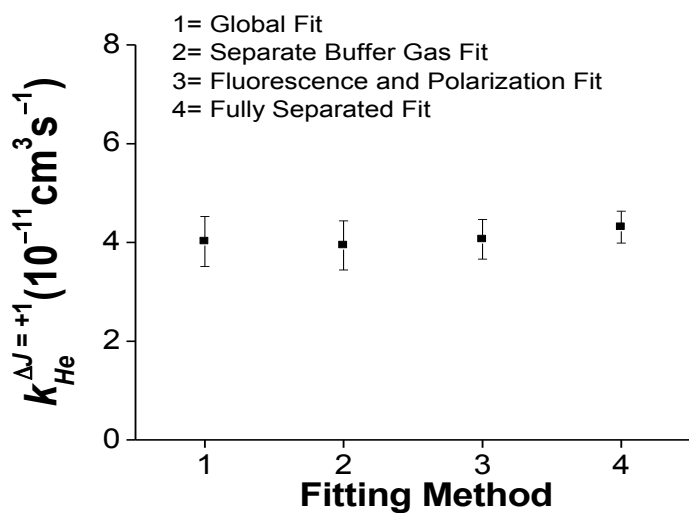
(c)  $\Delta J = -3$



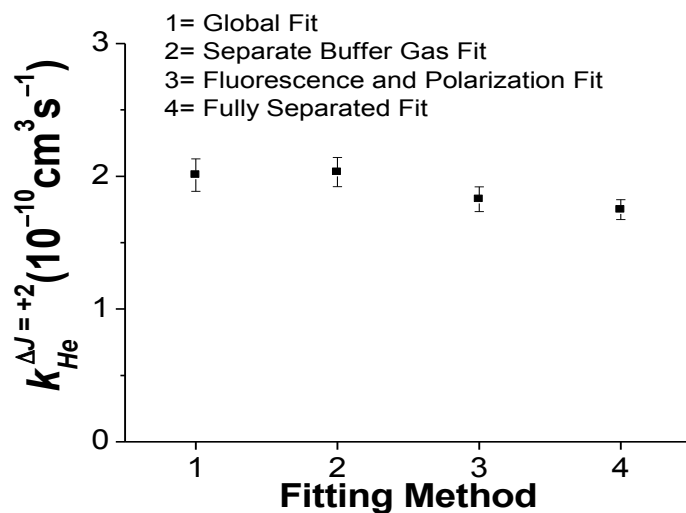
(d)  $\Delta J = -4$

Supplementary Material Fig. 1b: Plots of the fitted argon  $J$ -changing rate coefficients  $k_{Ar}^{\Delta J}$  following excitation of NaK molecules to the  $2(A)^1\Sigma^+(v=16, J=30)$  level, for each of the fitting methods described in the text ( $-4 \leq \Delta J \leq -1$ ).

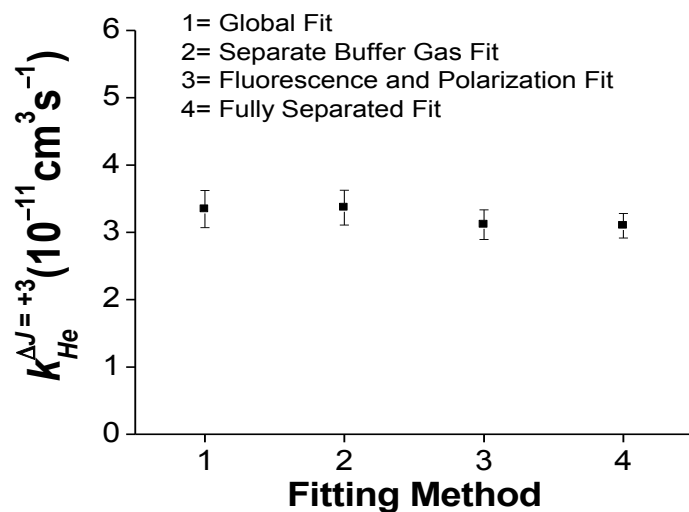
# $k_{He}^{\Delta J}$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$



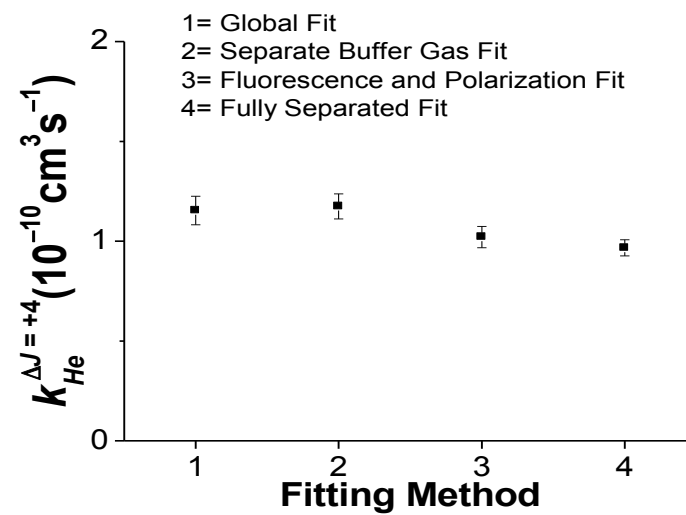
(a)  $\Delta J = +1$



(b)  $\Delta J = +2$



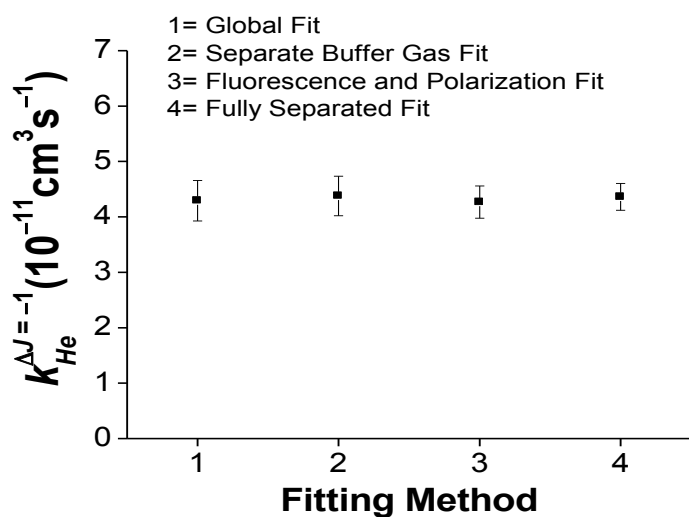
(c)  $\Delta J = +3$



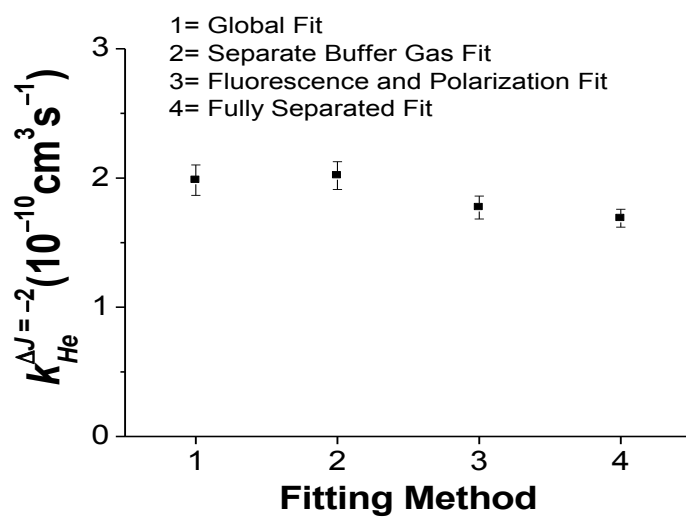
(d)  $\Delta J = +4$

Supplementary Material Fig. 2a: Plots of the fitted helium  $J$ -changing rate coefficients  $k_{He}^{\Delta J}$  following excitation of NaK molecules to the  $2(A)^1\Sigma^+(v=16, J=30)$  level, for each of the fitting methods described in the text ( $+4 \geq \Delta J \geq +1$ ).

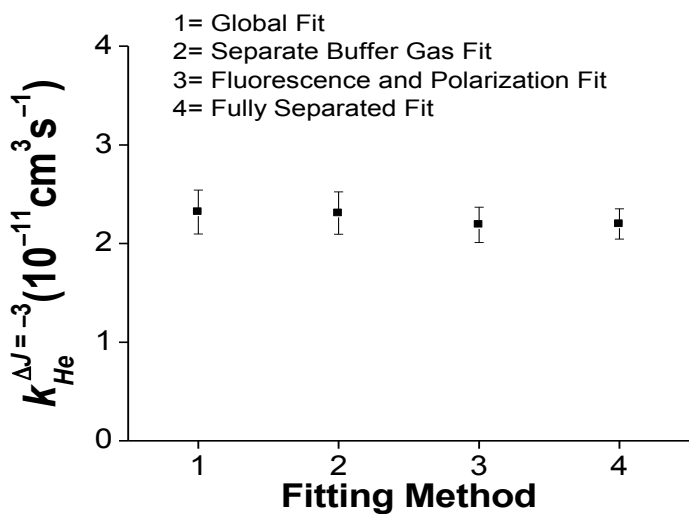
# $k_{He}^{\Delta J}$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$



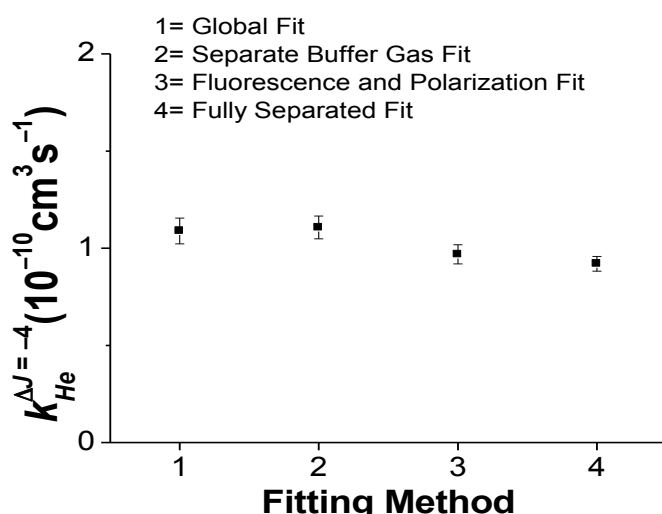
(a)  $\Delta J = -1$



(b)  $\Delta J = -2$



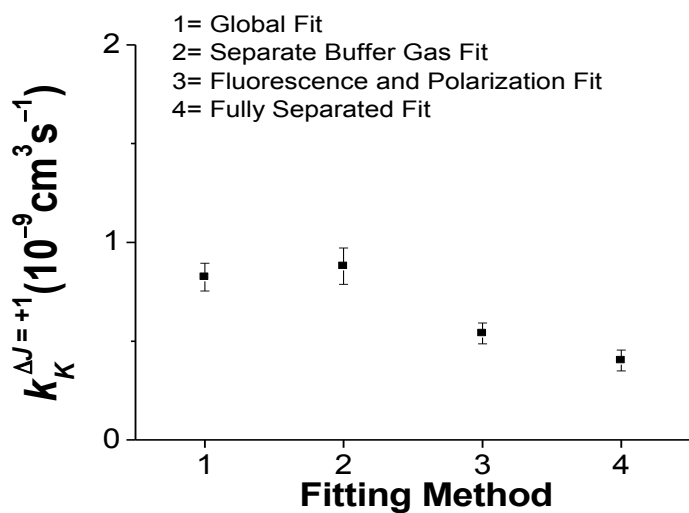
(c)  $\Delta J = -3$



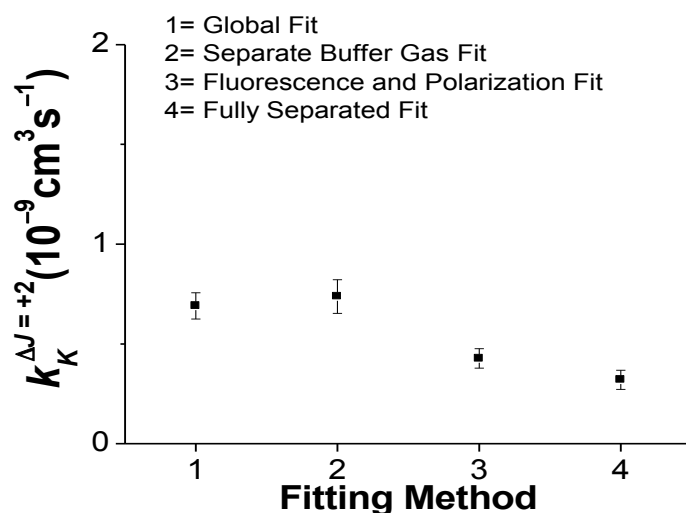
(d)  $\Delta J = -4$

Supplementary Material Fig. 2b: Plots of the fitted helium  $J$ -changing rate coefficients  $k_{He}^{\Delta J}$  following excitation of NaK molecules to the  $2(A)^1\Sigma^+(v=16, J=30)$  level, for each of the fitting methods described in the text ( $-4 \leq \Delta J \leq -1$ ).

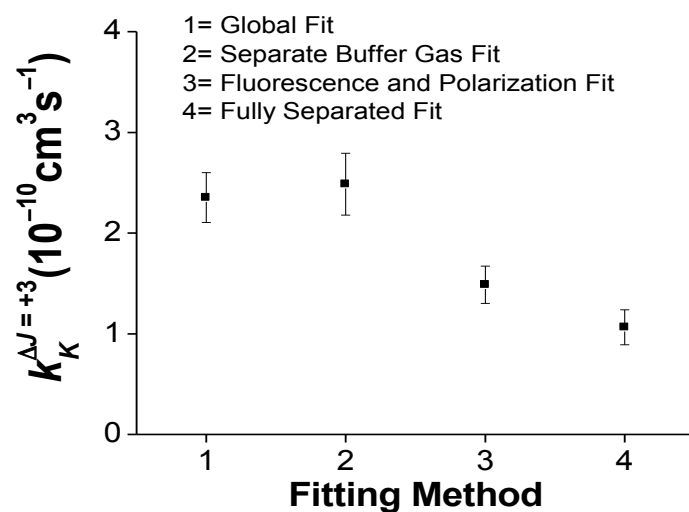
# $k_K^{\Delta J}$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$



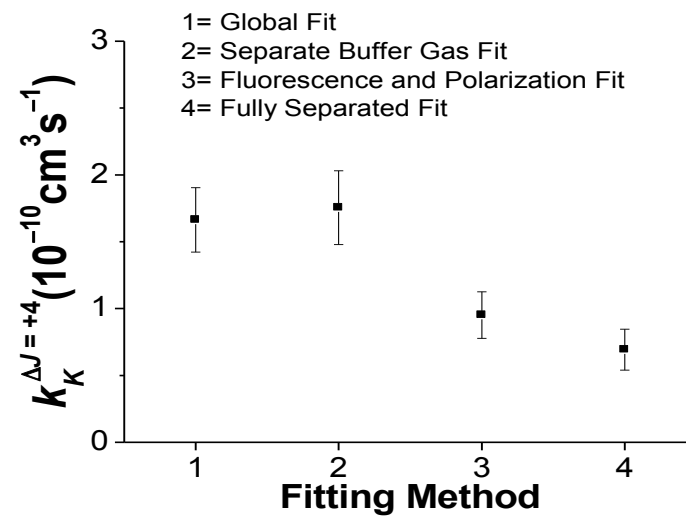
(a)  $\Delta J = +1$



(b)  $\Delta J = +2$



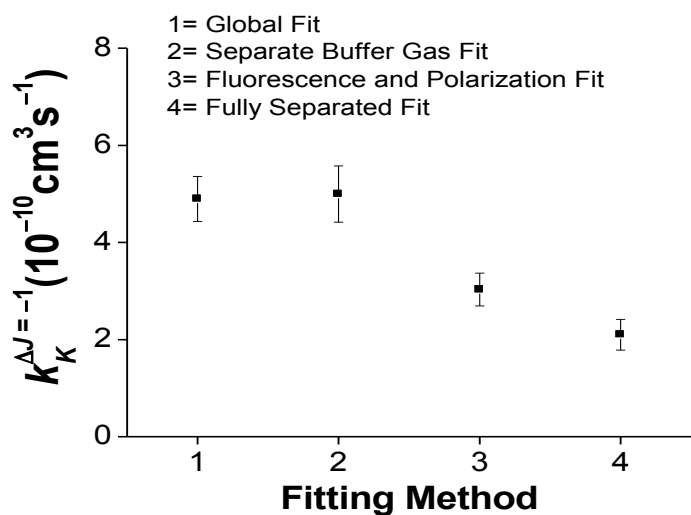
(c)  $\Delta J = +3$



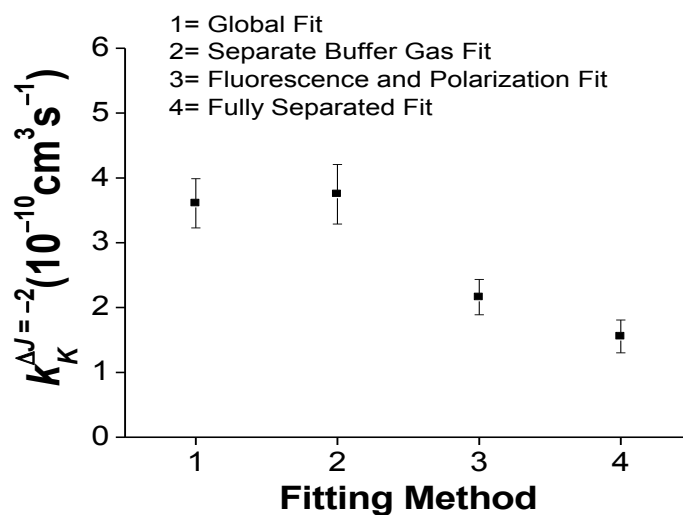
(d)  $\Delta J = +4$

Supplementary Material Fig. 3a: Plots of the fitted potassium  $J$ -changing rate coefficients  $k_K^{\Delta J}$  following excitation of NaK molecules to the  $2(A)^1\Sigma^+(v=16, J=30)$  level, for each of the fitting methods described in the text ( $+4 \geq \Delta J \geq +1$ ).

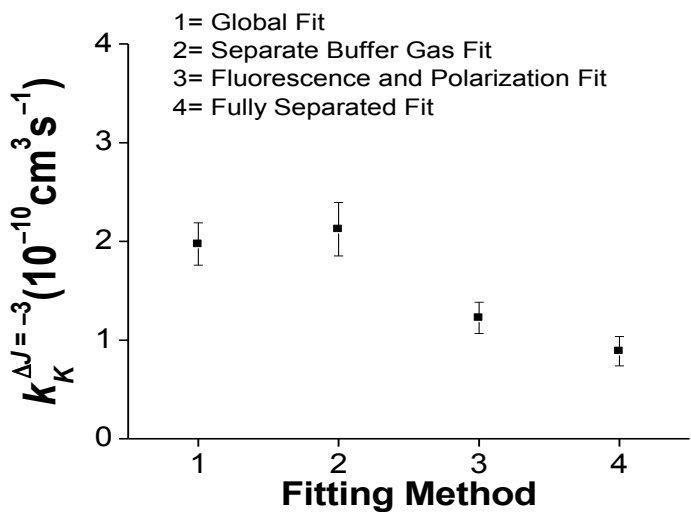
# $k_K^{\Delta J}$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$



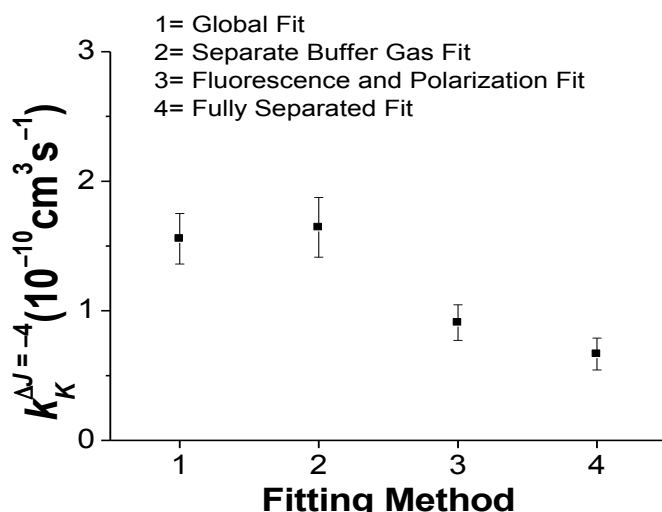
(a)  $\Delta J = -1$



(b)  $\Delta J = -2$



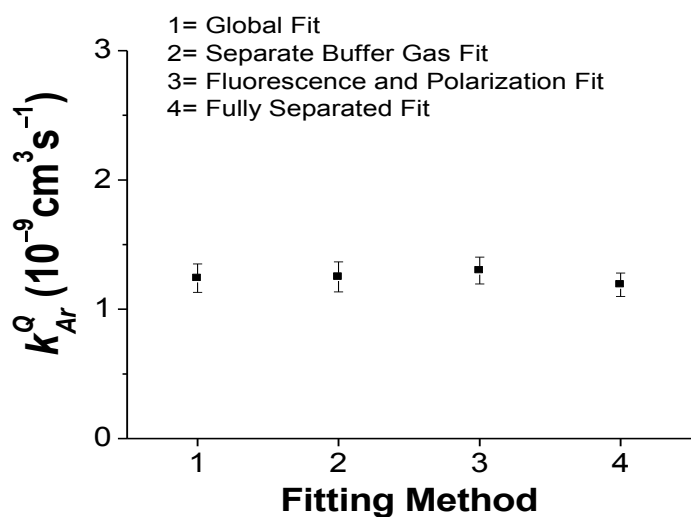
(c)  $\Delta J = -3$



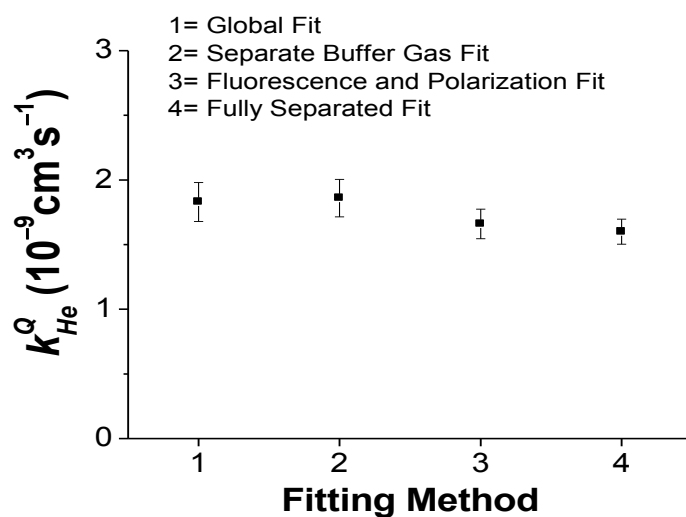
(d)  $\Delta J = -4$

Supplementary Material Fig. 3b: Plots of the fitted potassium  $J$ -changing rate coefficients  $k_K^{\Delta J}$  following excitation of NaK molecules to the  $2(A)^1\Sigma^+(v=16, J=30)$  level, for each of the fitting methods described in the text ( $-4 \leq \Delta J \leq -1$ ).

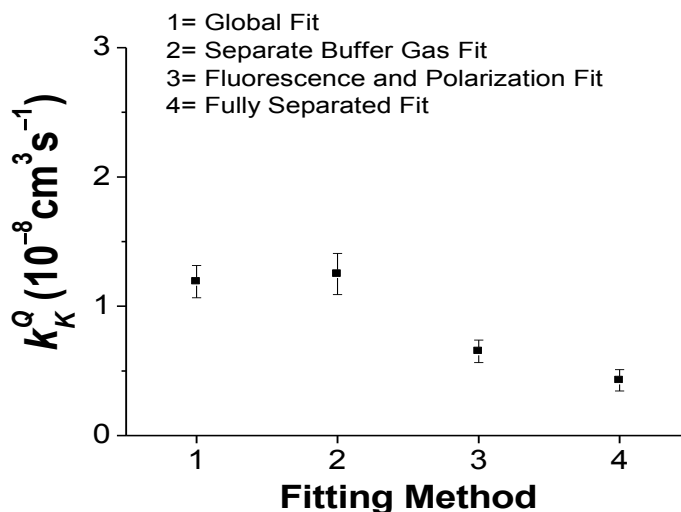
# $k_p^Q$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$



(a)  $k_{Ar}^Q$



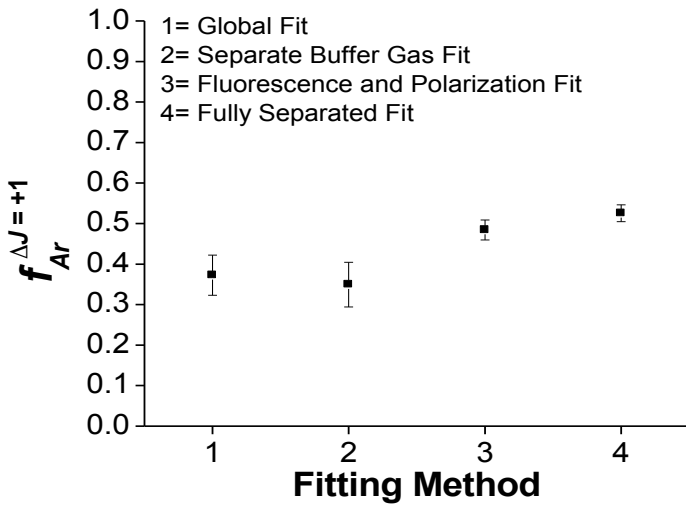
(b)  $k_{He}^Q$



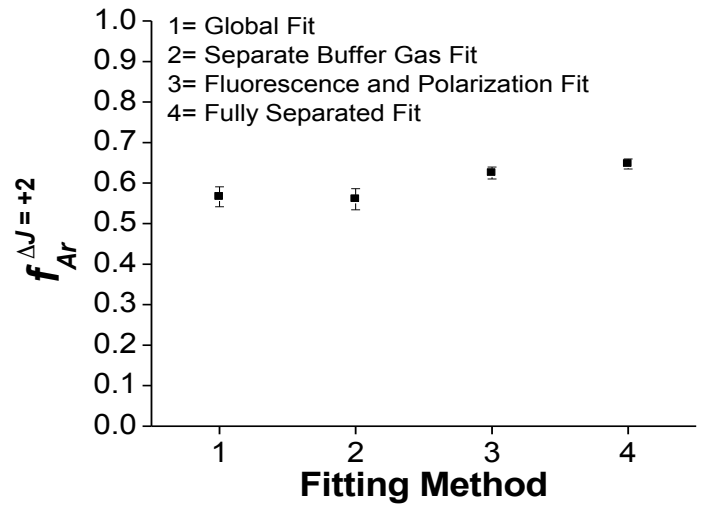
(c)  $k_K^Q$

Supplementary Material Fig. 4: Plots of the fitted quenching rate coefficients  $k_p^Q$  following excitation of NaK molecules to the  $2(A)^1\Sigma^+(v=16, J=30)$  level, for each of the fitting methods described in the text. a) argon, b) helium, and c) potassium collisions.

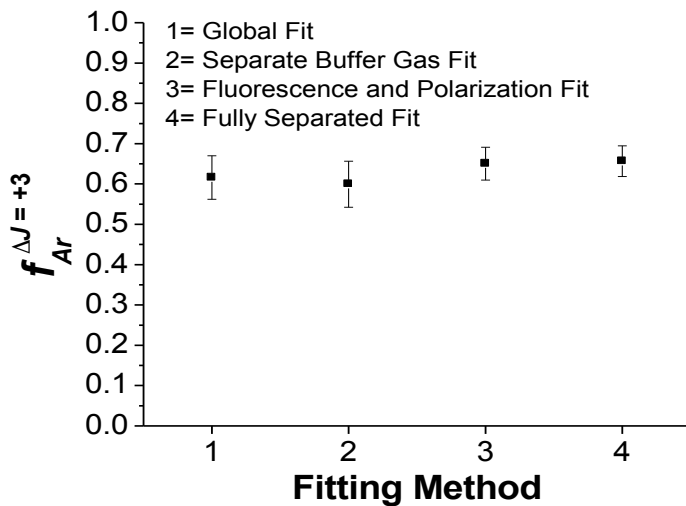
# $f_{Ar}^{\Delta J}$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$



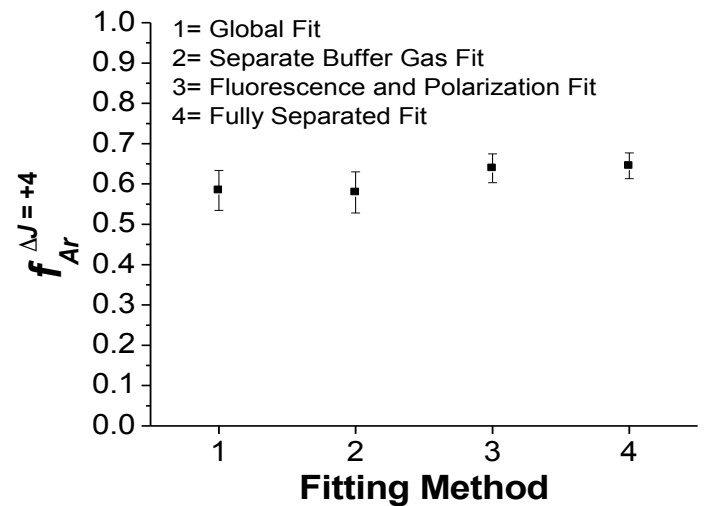
(a)  $\Delta J = +1$



(b)  $\Delta J = +2$



(c)  $\Delta J = +3$

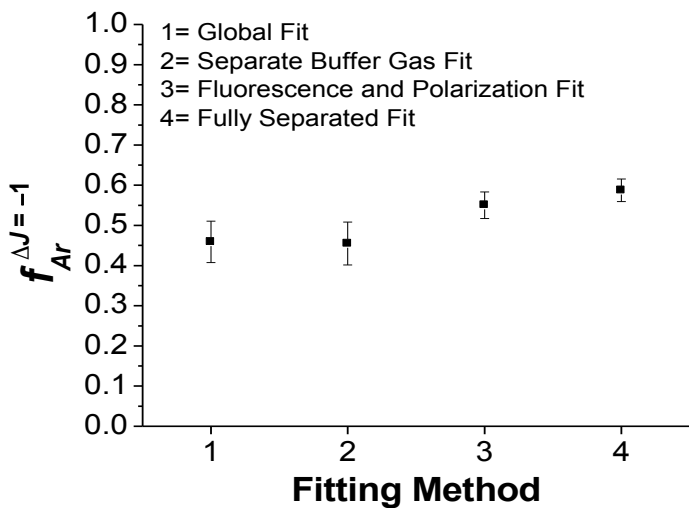


(d)  $\Delta J = +4$

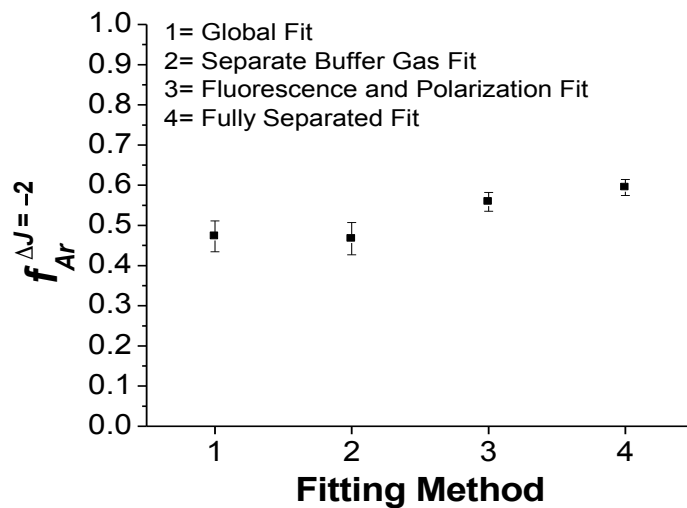
Supplementary Material Fig. 5a: Plots of the fitted fraction of orientation destroyed  $f_{Ar}^{\Delta J}$  for argon collisions following excitation of NaK molecules to the  $2(A)^1\Sigma^+(v=16, J=30)$  level, for each of the fitting methods described in the text ( $+4 \geq \Delta J \geq +1$ ).



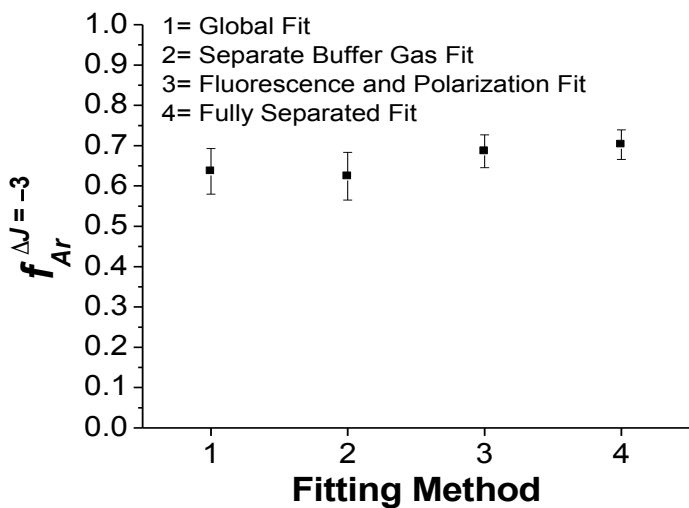
# $f_{Ar}^{\Delta J}$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$



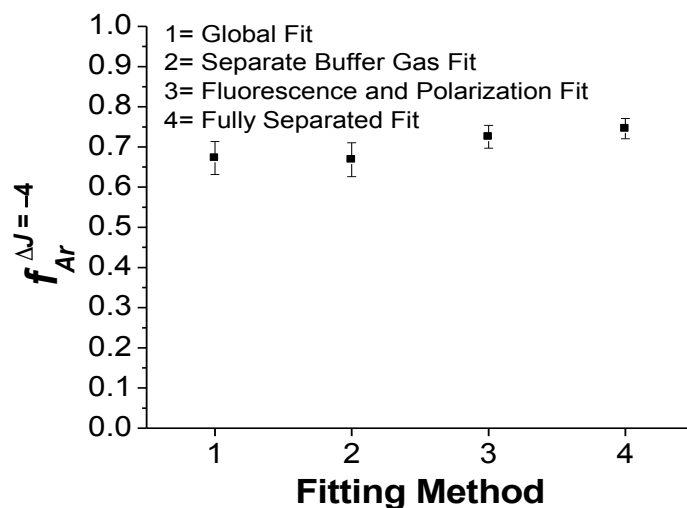
(a)  $\Delta J = -1$



(b)  $\Delta J = -2$



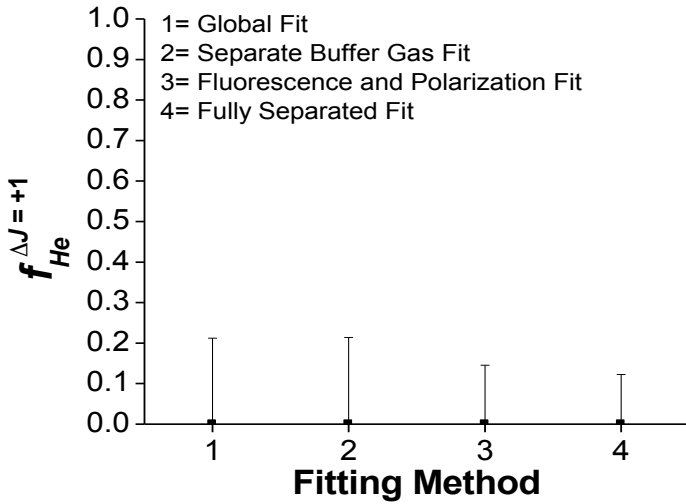
(c)  $\Delta J = -3$



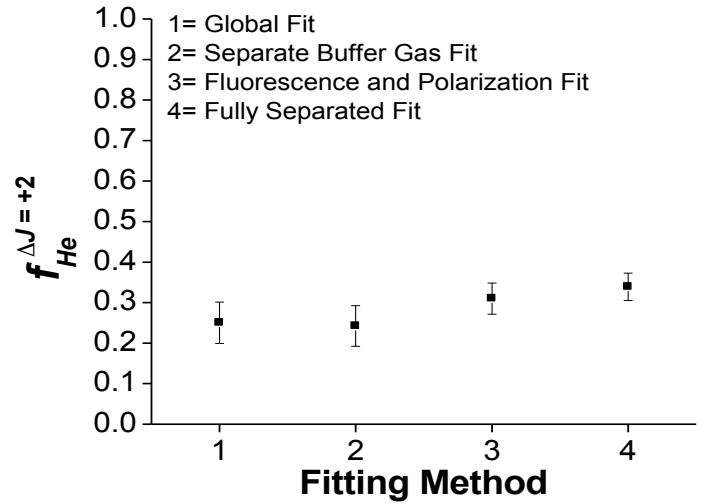
(d)  $\Delta J = -4$

Supplementary Material Fig. 5b: Plots of the fitted fraction of orientation destroyed  $f_{Ar}^{\Delta J}$  for argon collisions following excitation of NaK molecules to the  $2(A)^1\Sigma^+(v=16, J=30)$  level, for each of the fitting methods described in the text ( $-4 \leq \Delta J \leq -1$ ).

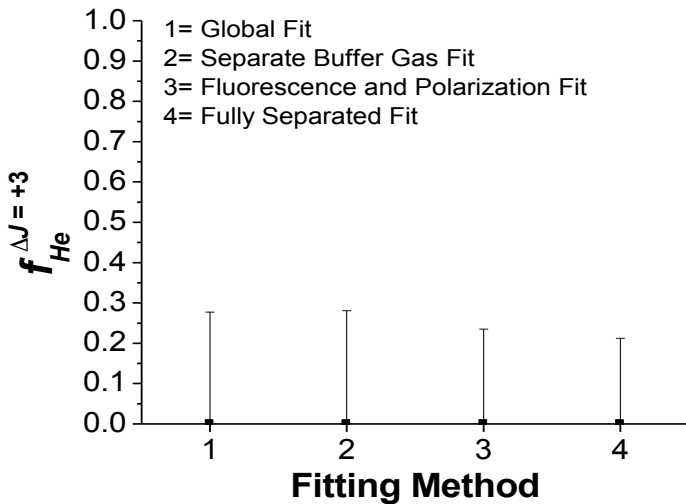
# $f_{He}^{\Delta J}$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$



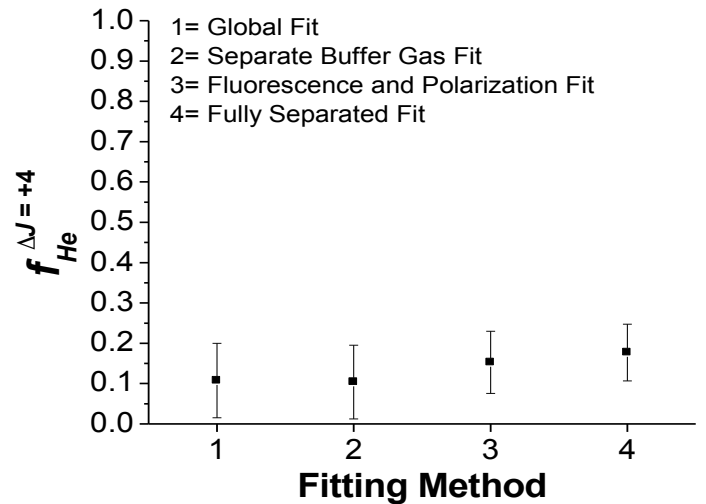
(a)  $\Delta J = +1$



(b)  $\Delta J = +2$



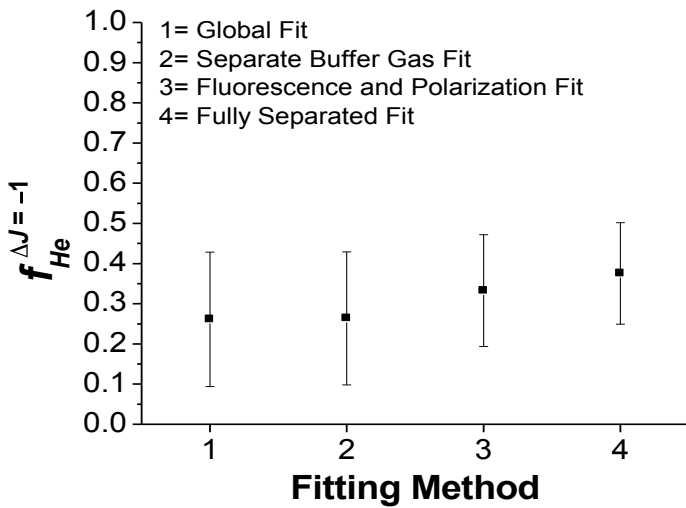
(c)  $\Delta J = +3$



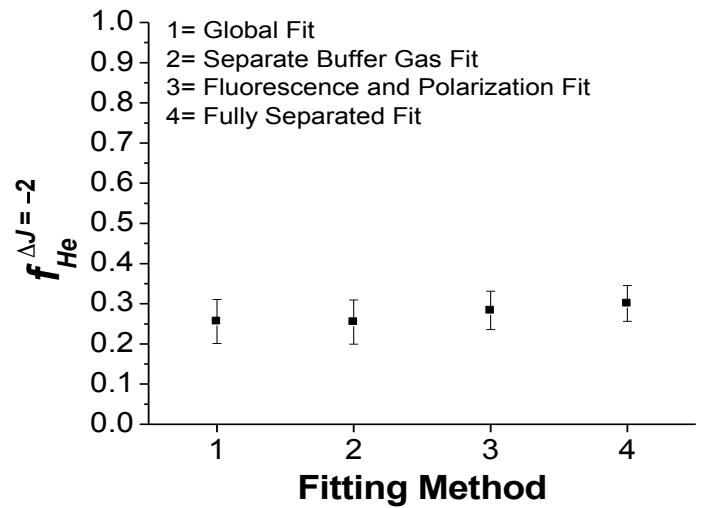
(d)  $\Delta J = +4$

Supplementary Material Fig. 6a: Plots of the fitted fraction of orientation destroyed  $f_{He}^{\Delta J}$  for helium collisions following excitation of NaK molecules to the  $2(A)^1\Sigma^+(v=16, J=30)$  level, for each of the fitting methods described in the text ( $+4 \geq \Delta J \geq +1$ ).

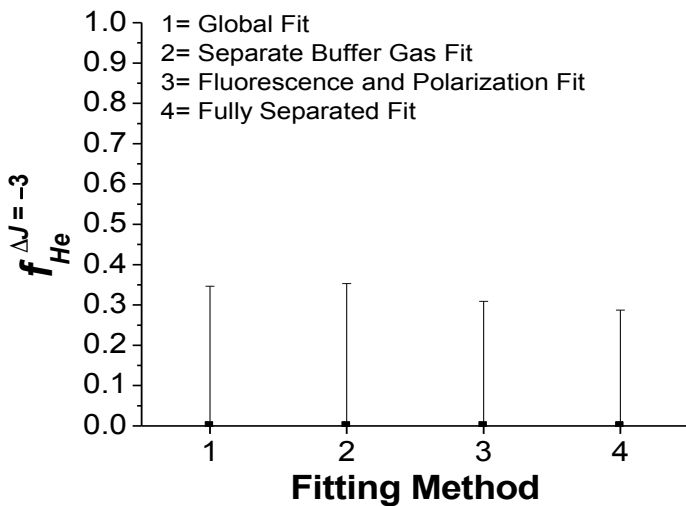
# $f_{He}^{\Delta J}$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$



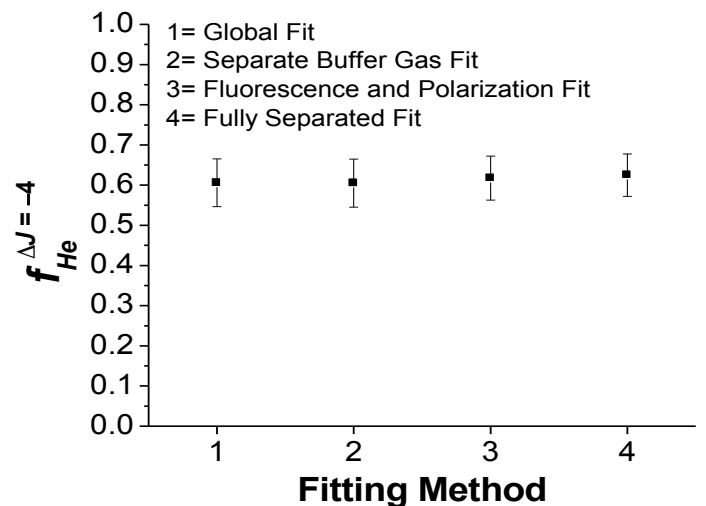
(a)  $\Delta J = -1$



(b)  $\Delta J = -2$



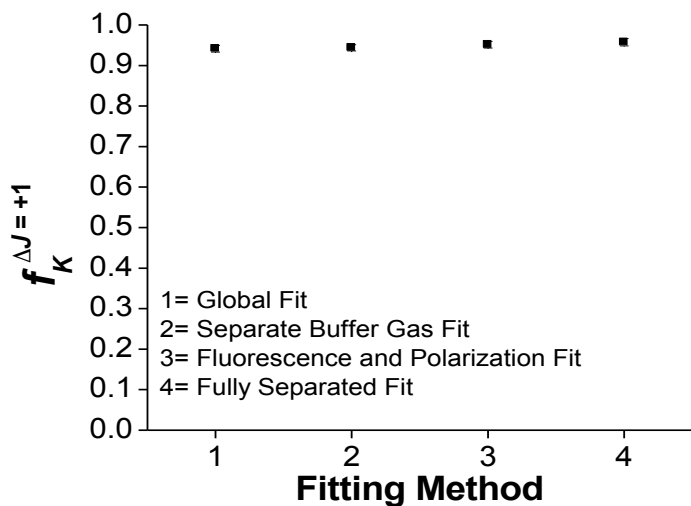
(c)  $\Delta J = -3$



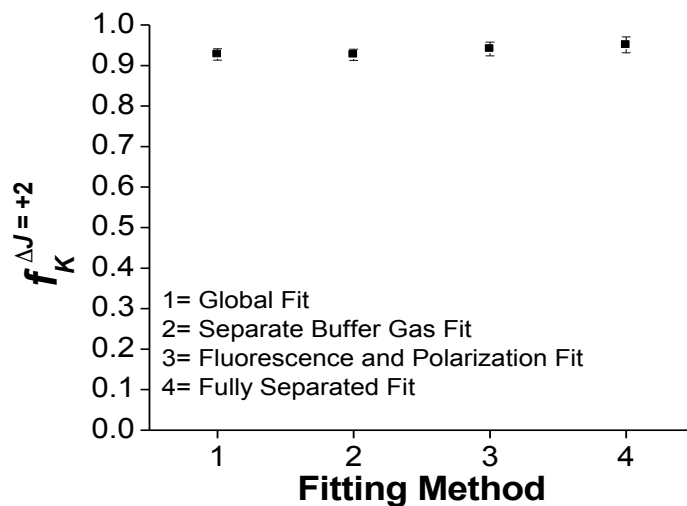
(d)  $\Delta J = -4$

Supplementary Material Fig. 6b: Plots of the fitted fraction of orientation destroyed  $f_{He}^{\Delta J}$  for helium collisions following excitation of NaK molecules to the  $2(A)^1\Sigma^+(v=16, J=30)$  level, for each of the fitting methods described in the text ( $-4 \leq \Delta J \leq -1$ ).

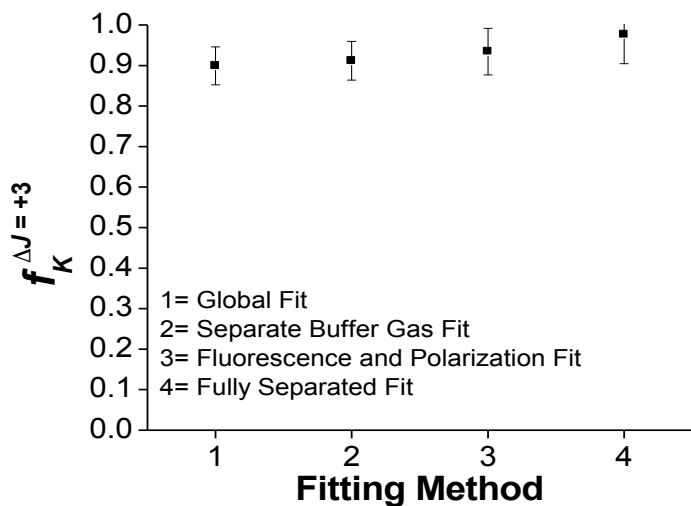
# $f_K^{\Delta J}$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$



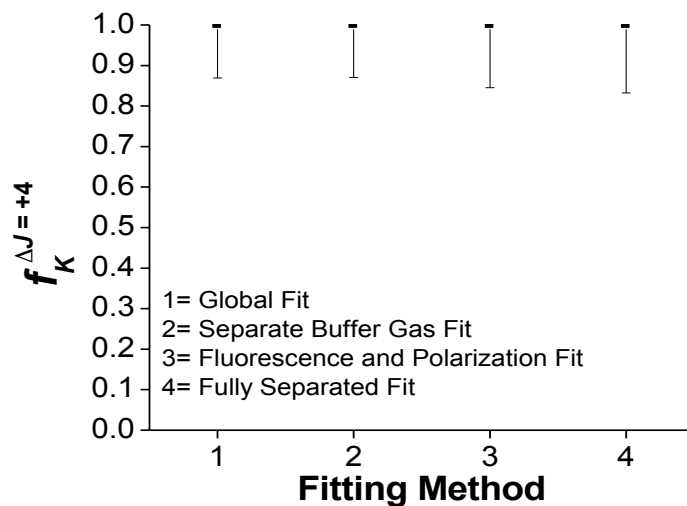
(a)  $\Delta J = +1$



(b)  $\Delta J = +2$



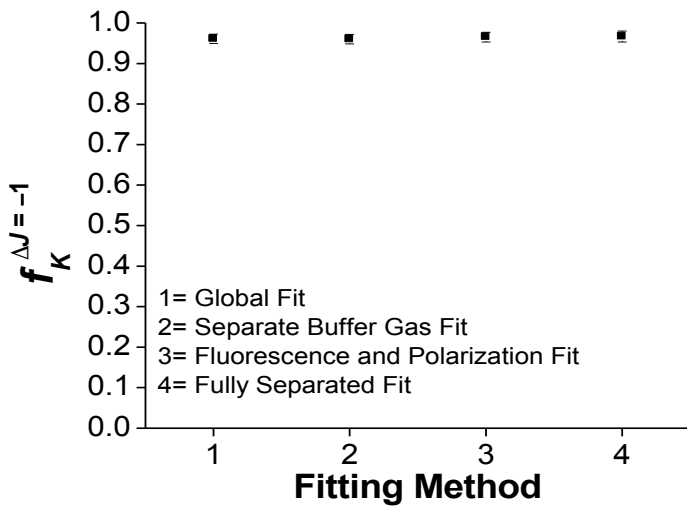
(c)  $\Delta J = +3$



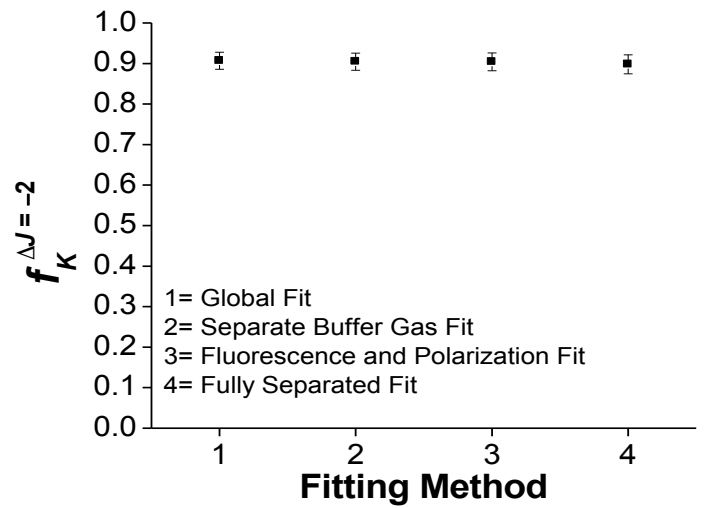
(d)  $\Delta J = +4$

Supplementary Material Fig. 7a: Plots of the fitted fraction of orientation destroyed  $f_K^{\Delta J}$  for potassium collisions following excitation of NaK molecules to the  $2(A)^1\Sigma^+(v=16, J=30)$  level, for each of the fitting methods described in the text ( $+4 \geq \Delta J \geq +1$ ).

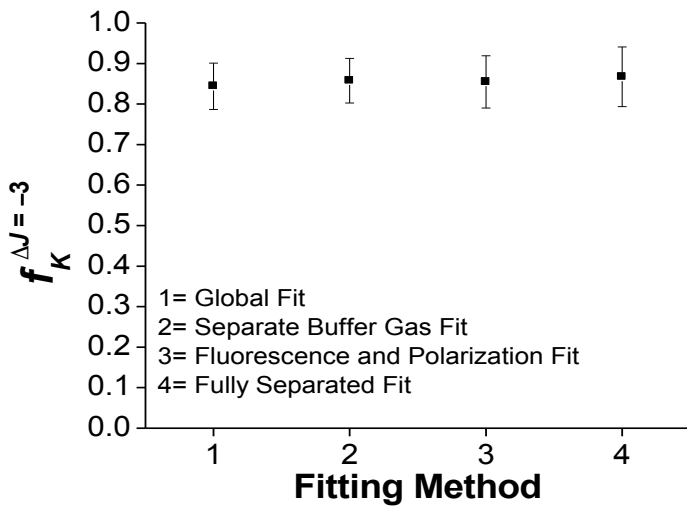
# $f_K^{\Delta J}$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$



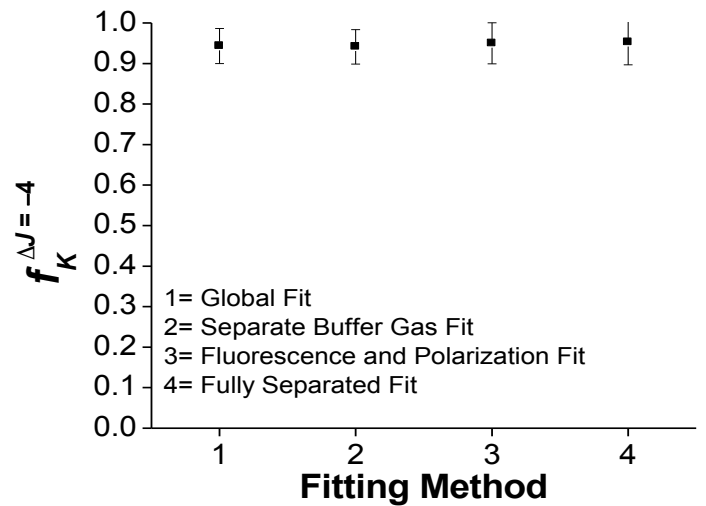
(a)  $\Delta J = -1$



(b)  $\Delta J = -2$



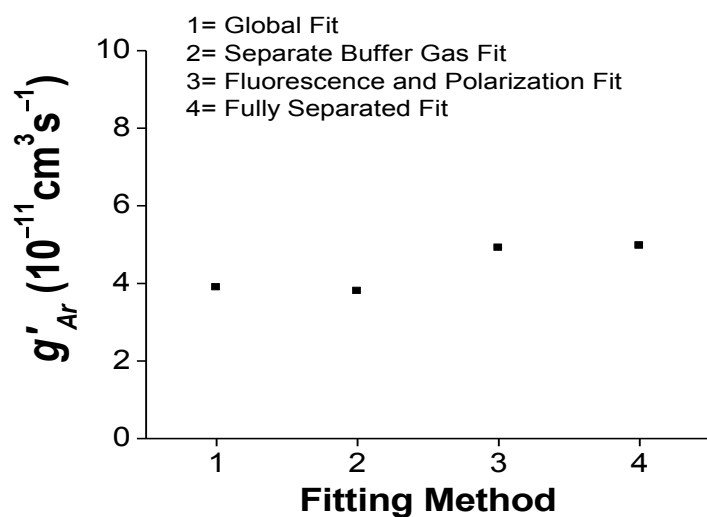
(c)  $\Delta J = -3$



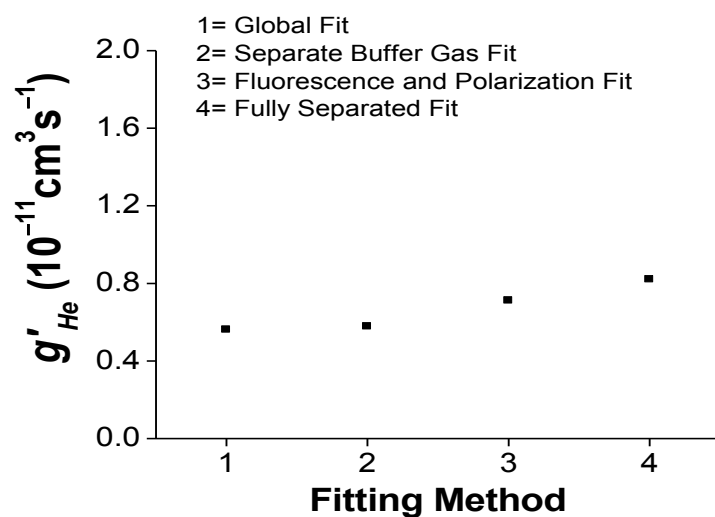
(d)  $\Delta J = -4$

Supplementary Material Fig. 7b: Plots of the fitted fraction of orientation destroyed  $f_K^{\Delta J}$  for potassium collisions following excitation of NaK molecules to the  $2(A)^1\Sigma^+(v=16, J=30)$  level, for each of the fitting methods described in the text ( $-4 \leq \Delta J \leq -1$ ).

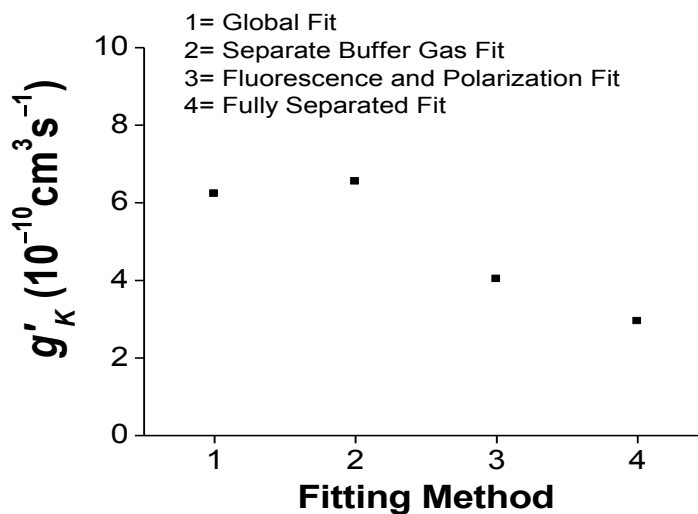
# $g'_p$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$



(a)  $g'_{Ar}$



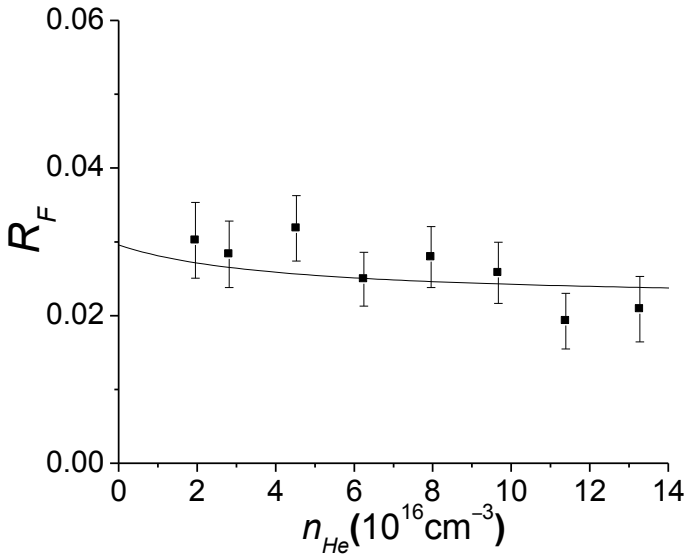
(b)  $g'_{He}$



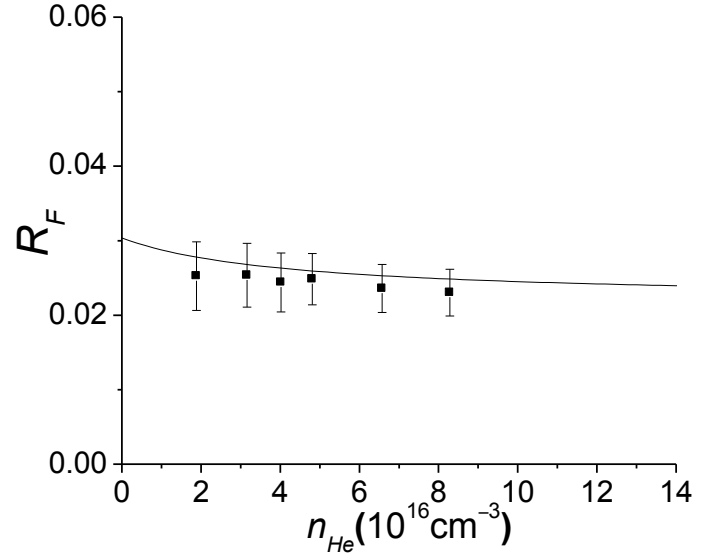
(c)  $g'_K$

Supplementary Material Fig. 8: Plots of the fitted rate coefficients for total destruction of orientation  $g'_p$  following excitation of NaK molecules to the  $2(A)^1\Sigma^+(v=16, J=30)$  level, for each of the fitting methods described in the text. a) argon, b) helium, and c) potassium collisions.

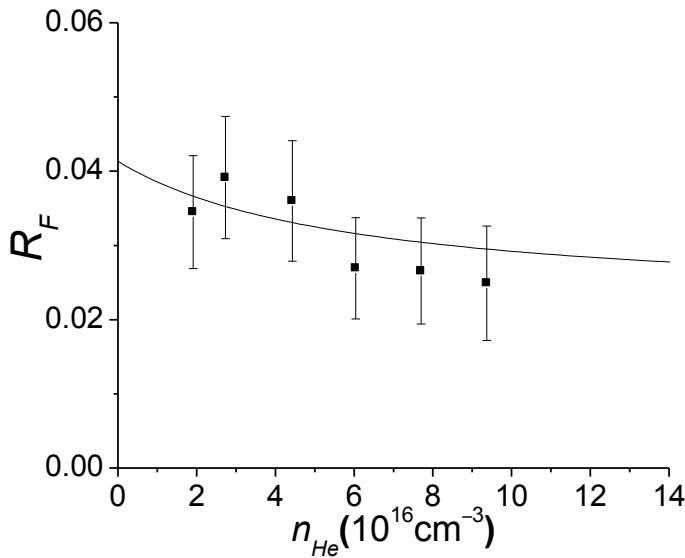
$R_F$  vs.  $n_{He}$  for NaK  $2(A)^1\Sigma^+(v=16, J=30)$ ,  $\Delta J = +1$



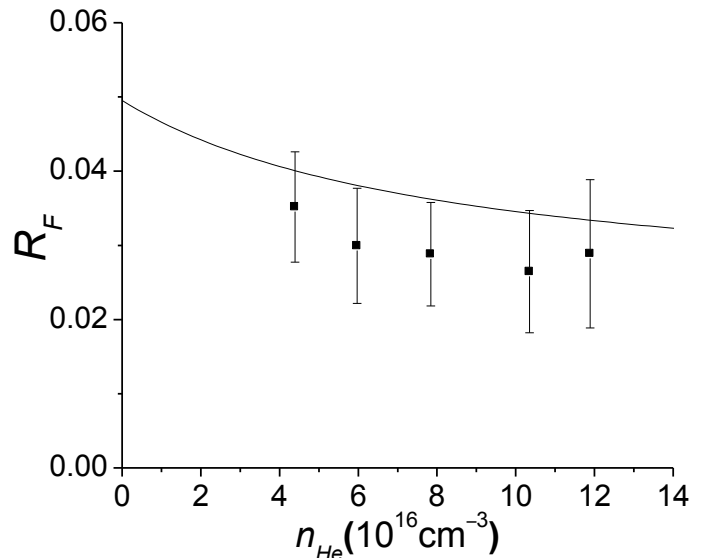
(a)  $n_K = 2.75 \times 10^{15} \text{ cm}^{-3}$



(b)  $n_K = 2.88 \times 10^{15} \text{ cm}^{-3}$



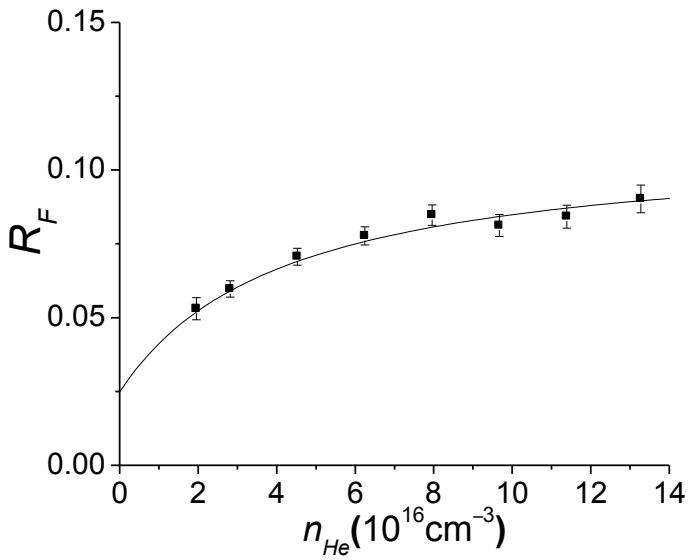
(c)  $n_K = 5.44 \times 10^{15} \text{ cm}^{-3}$



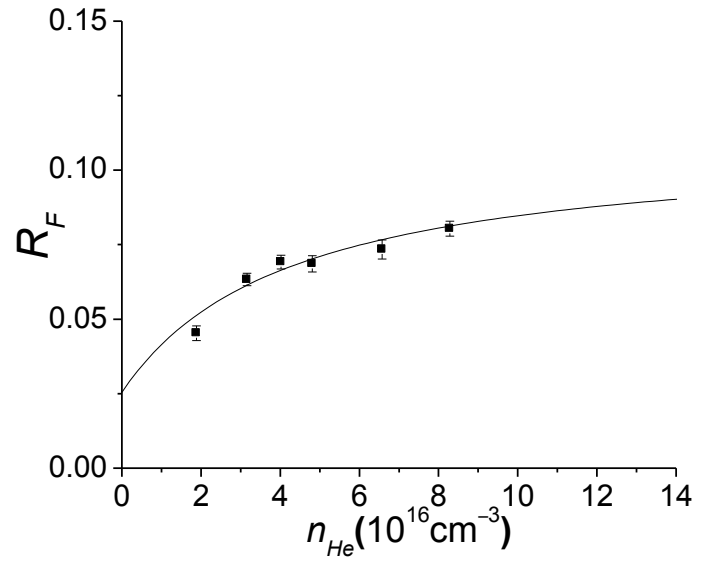
(d)  $n_K = 9.20 \times 10^{15} \text{ cm}^{-3}$

Supplementary Material Fig. 9: Plots of fluorescence ratio data ( $R_F$ ) versus helium density for  $\Delta J = +1$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ . In this, and subsequent plots, the calculated  $R_F$  values (solid curves) were obtained using the global fit parameters given in Table 1 of the paper (also listed in Supplementary Table 1).

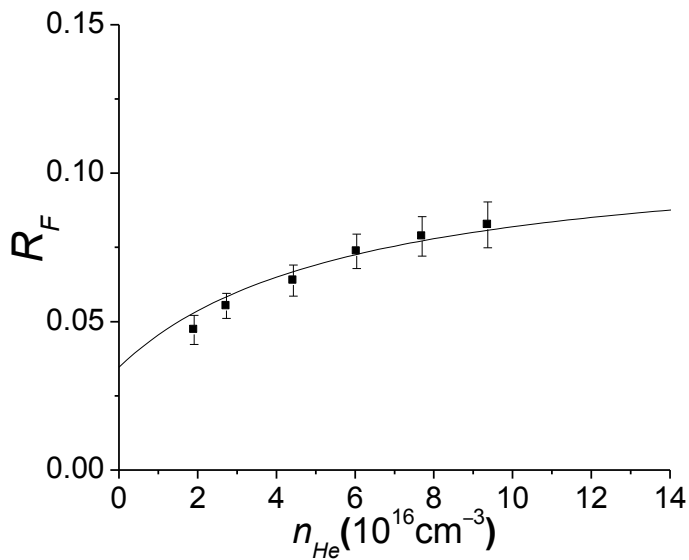
$R_F$  vs.  $n_{He}$  for NaK  $2(A)^1\Sigma^+(v=16, J=30)$ ,  $\Delta J = +2$



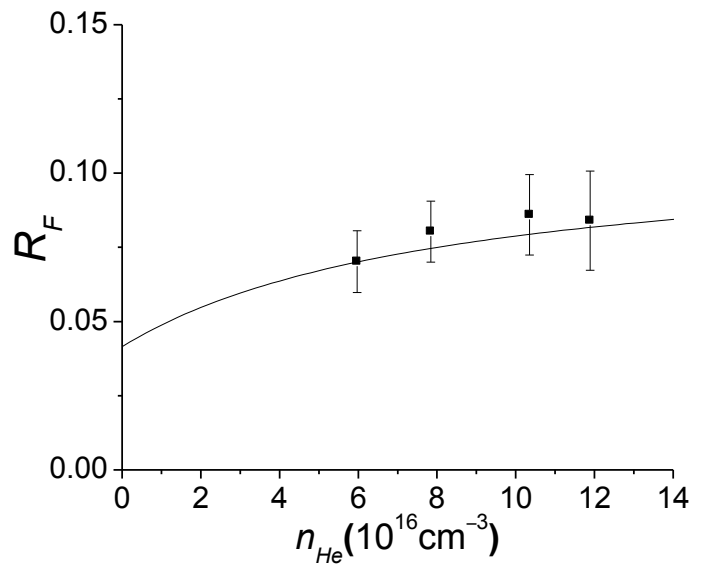
(a)  $n_K = 2.75 \times 10^{15} \text{ cm}^{-3}$



(b)  $n_K = 2.88 \times 10^{15} \text{ cm}^{-3}$



(c)  $n_K = 5.44 \times 10^{15} \text{ cm}^{-3}$

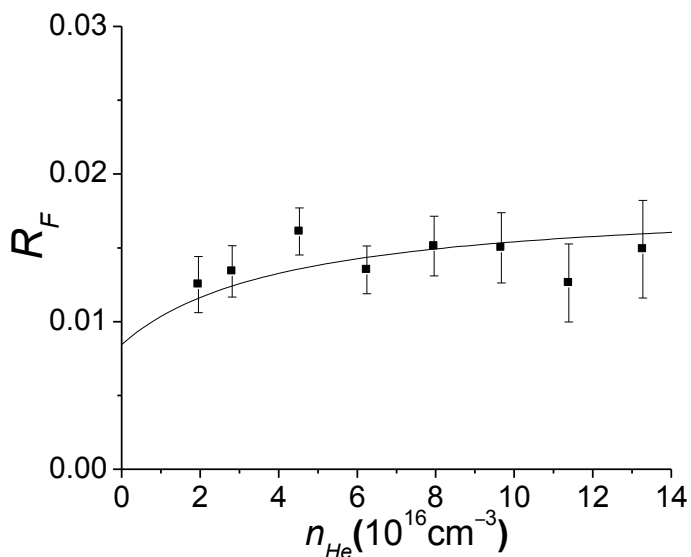


(d)  $n_K = 9.20 \times 10^{15} \text{ cm}^{-3}$

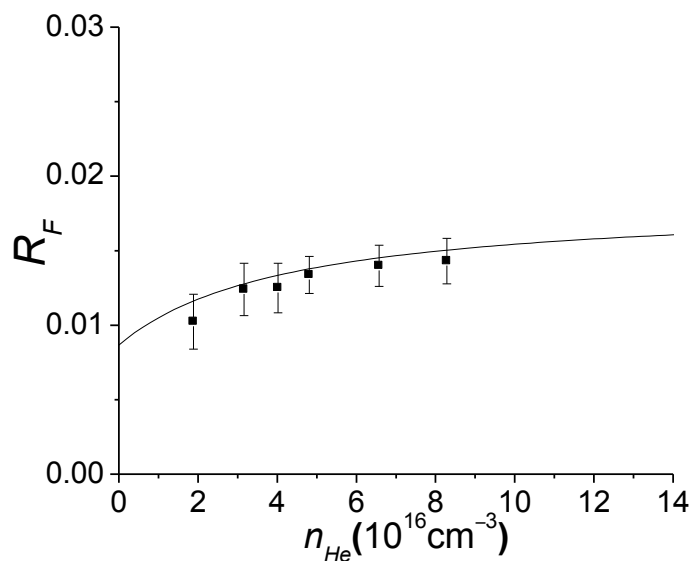
Supplementary Material Fig. 10: Plots of fluorescence ratio data ( $R_F$ ) versus helium density for  $\Delta J = +2$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ .



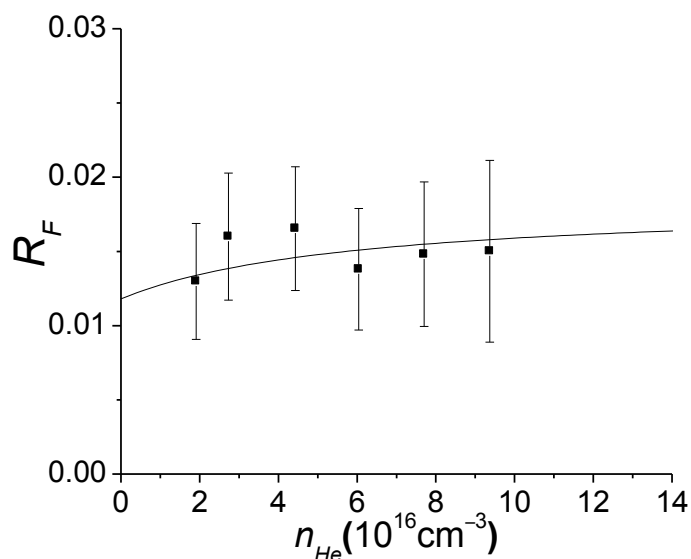
$R_F$  vs.  $n_{He}$  for NaK  $2(A)^1\Sigma^+(v=16, J=30)$ ,  $\Delta J = +3$



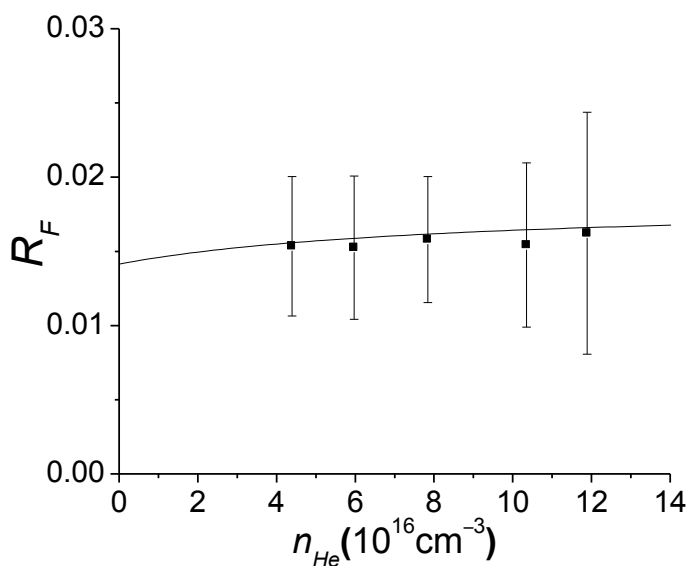
(a)  $n_K = 2.75 \times 10^{15} \text{ cm}^{-3}$



(b)  $n_K = 2.88 \times 10^{15} \text{ cm}^{-3}$



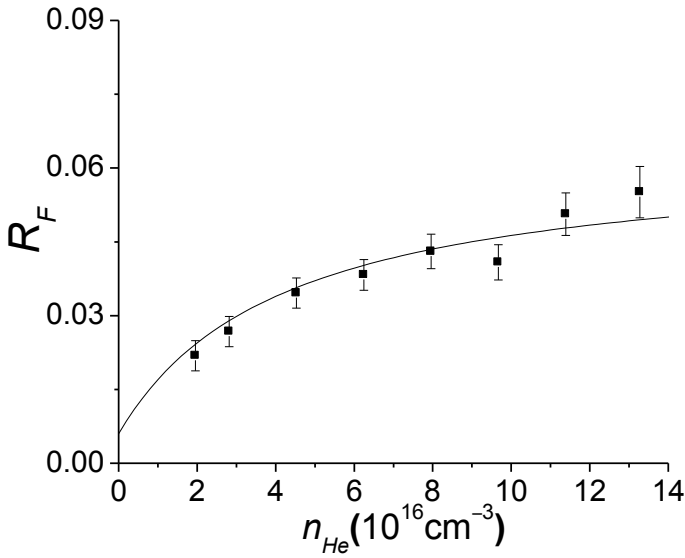
(c)  $n_K = 5.44 \times 10^{15} \text{ cm}^{-3}$



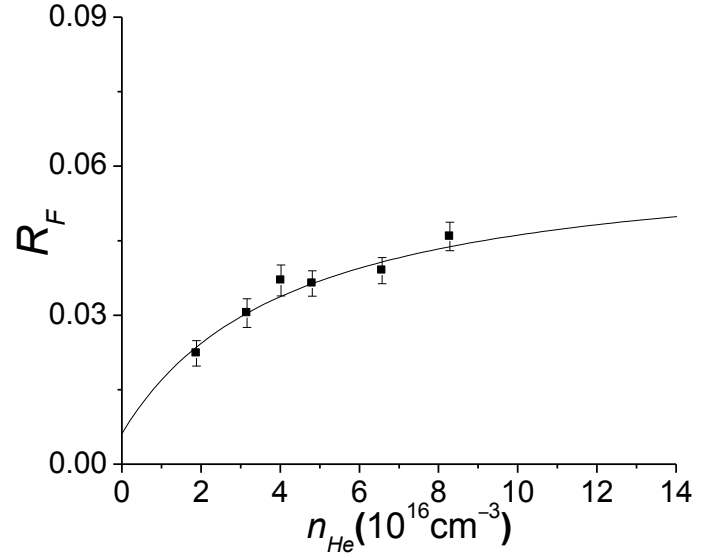
(d)  $n_K = 9.20 \times 10^{15} \text{ cm}^{-3}$

Supplementary Material Fig. 11: Plots of fluorescence ratio data ( $R_F$ ) versus helium density for  $\Delta J = +3$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ .

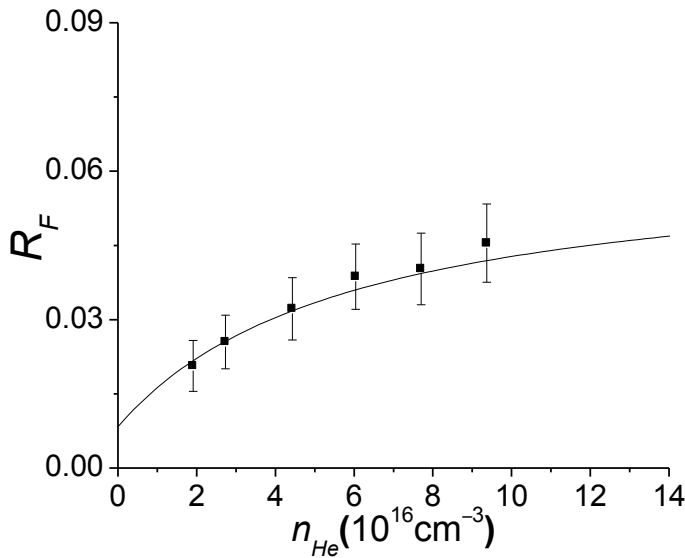
$R_F$  vs.  $n_{He}$  for NaK  $2(A)^1\Sigma^+(v=16, J=30)$ ,  $\Delta J = +4$



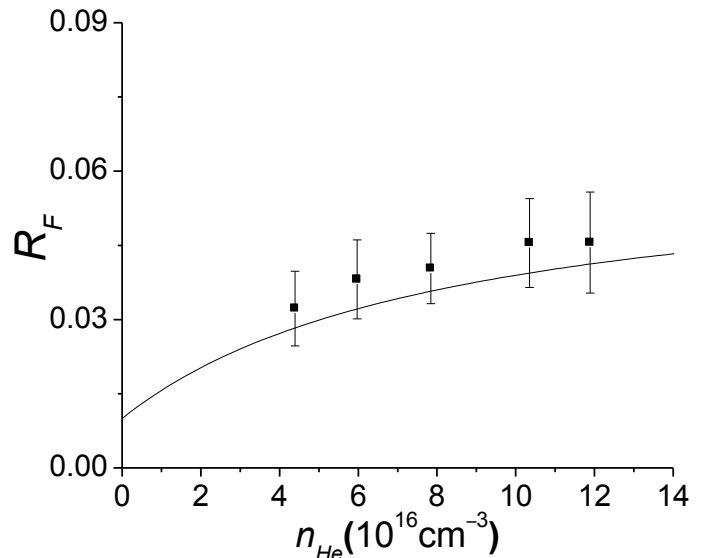
(a)  $n_K = 2.75 \times 10^{15} \text{ cm}^{-3}$



(b)  $n_K = 2.88 \times 10^{15} \text{ cm}^{-3}$



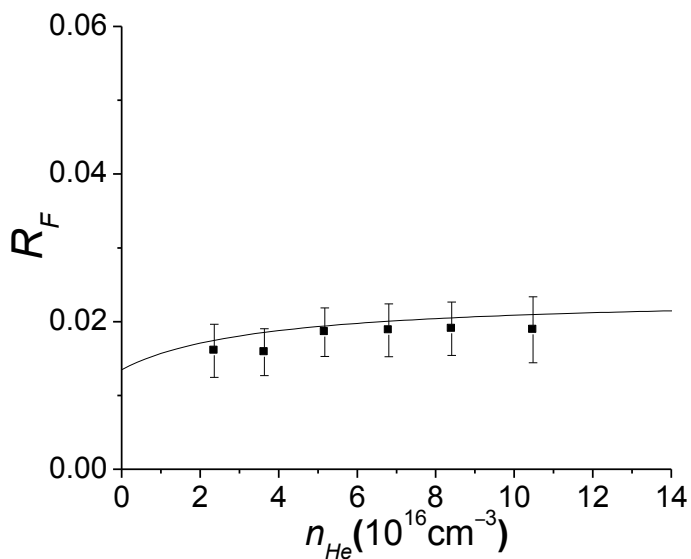
(c)  $n_K = 5.44 \times 10^{15} \text{ cm}^{-3}$



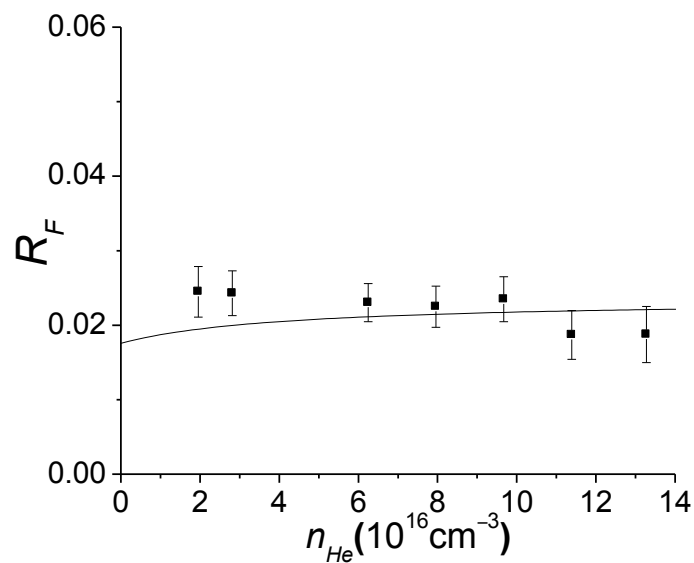
(d)  $n_K = 9.20 \times 10^{15} \text{ cm}^{-3}$

Supplementary Material Fig. 12: Plots of fluorescence ratio data ( $R_F$ ) versus helium density for  $\Delta J = +4$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ .

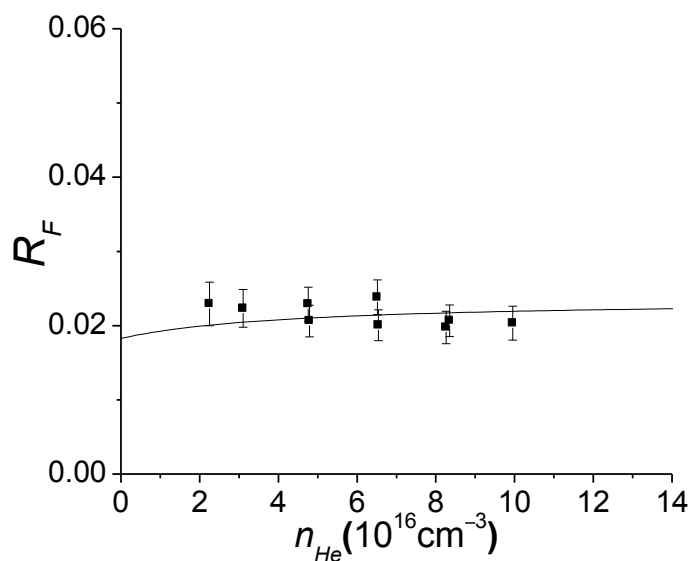
$R_F$  vs.  $n_{He}$  for NaK  $2(A)^1\Sigma^+(v=16, J=30), \Delta J = -1$



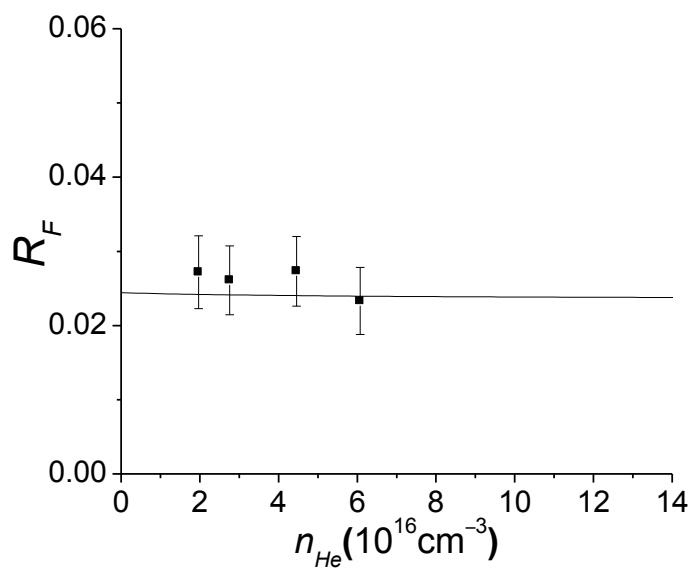
(a)  $n_K = 1.80 \times 10^{15} \text{ cm}^{-3}$



(b)  $n_K = 2.75 \times 10^{15} \text{ cm}^{-3}$



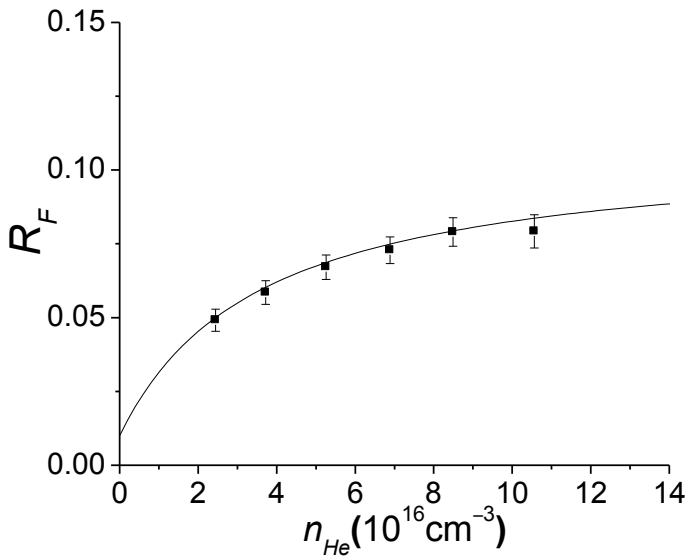
(c)  $n_K = 2.94 \times 10^{15} \text{ cm}^{-3}$



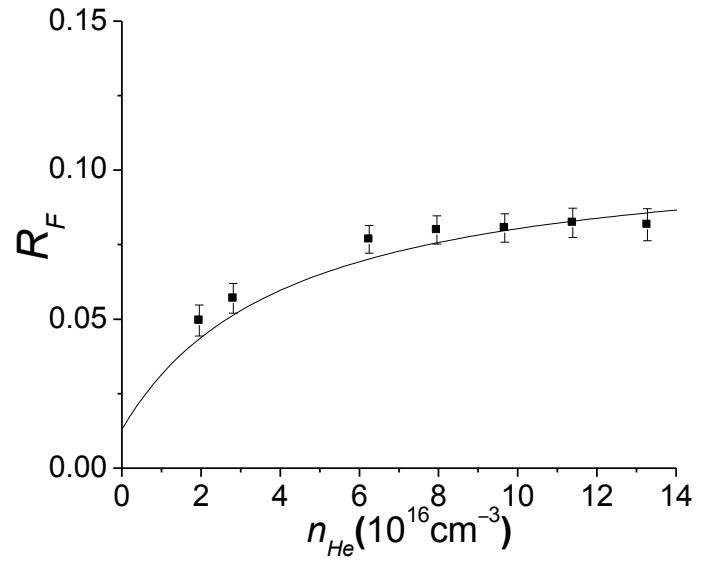
(d)  $n_K = 5.37 \times 10^{15} \text{ cm}^{-3}$

Supplementary Material Fig. 13: Plots of fluorescence ratio data ( $R_F$ ) versus helium density for  $\Delta J = -1$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ .

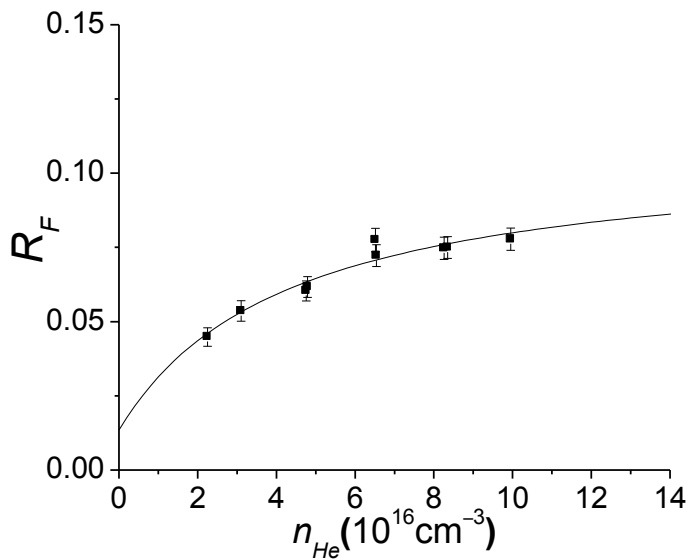
$R_F$  vs.  $n_{He}$  for NaK  $2(A)^1\Sigma^+(v=16, J=30)$ ,  $\Delta J = -2$



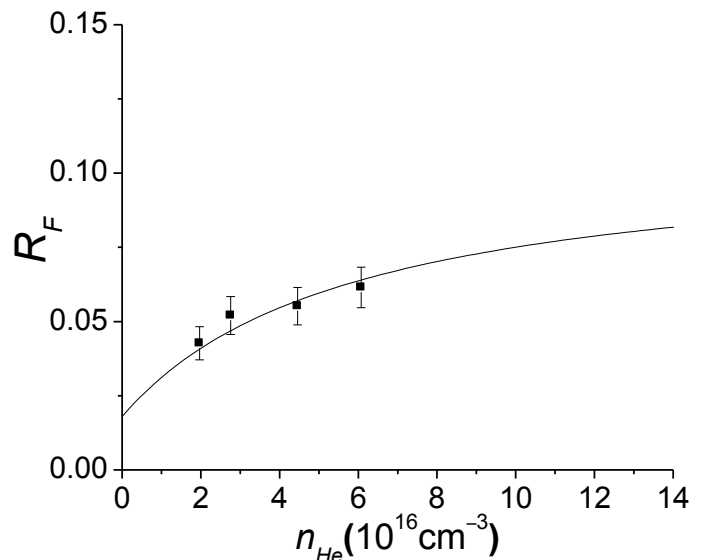
(a)  $n_K = 1.80 \times 10^{15} \text{ cm}^{-3}$



(b)  $n_K = 2.75 \times 10^{15} \text{ cm}^{-3}$



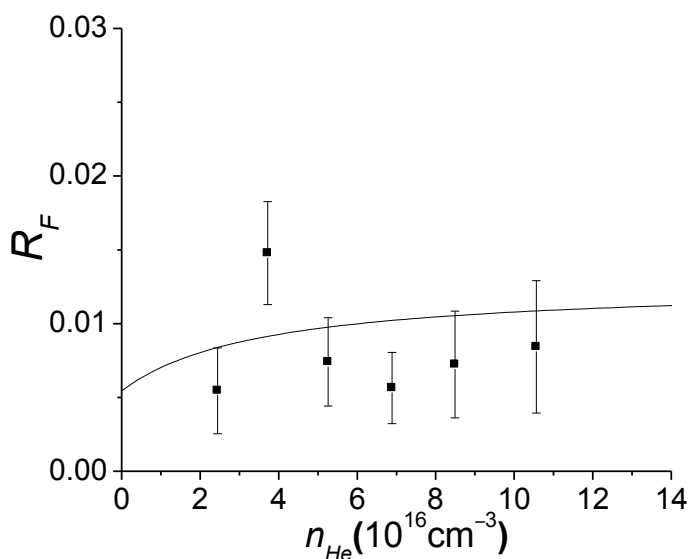
(c)  $n_K = 2.94 \times 10^{15} \text{ cm}^{-3}$



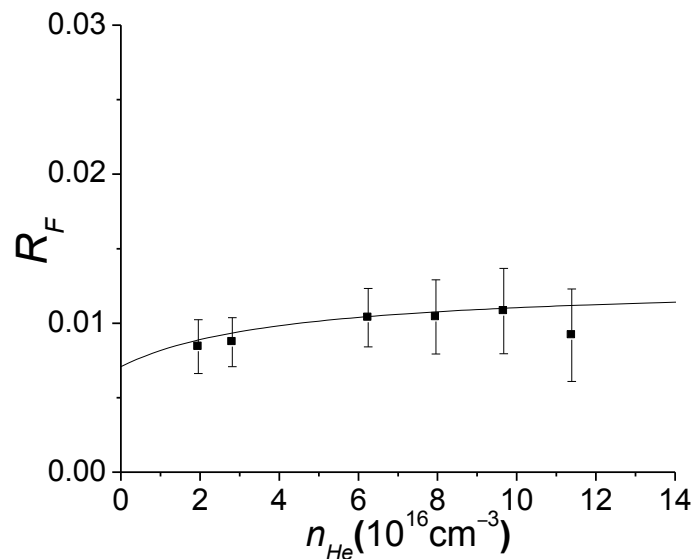
(d)  $n_K = 5.37 \times 10^{15} \text{ cm}^{-3}$

Supplementary Material Fig. 14: Plots of fluorescence ratio data ( $R_F$ ) versus helium density for  $\Delta J = -2$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ .

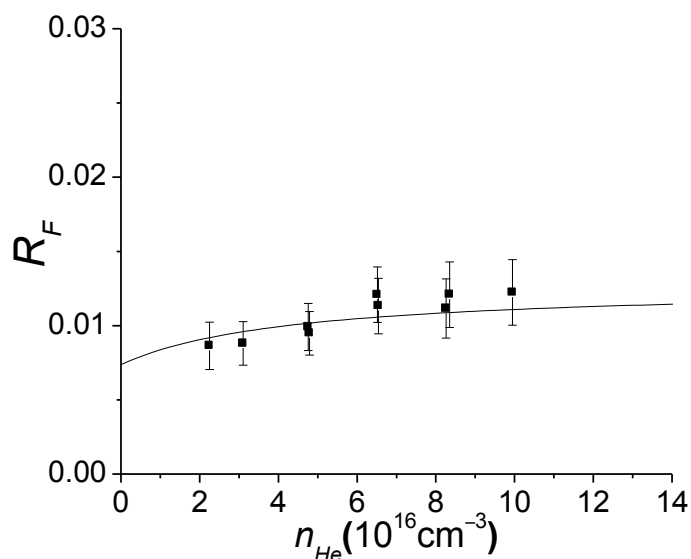
$R_F$  vs.  $n_{He}$  for NaK  $2(A)^1\Sigma^+(v=16, J=30)$ ,  $\Delta J = -3$



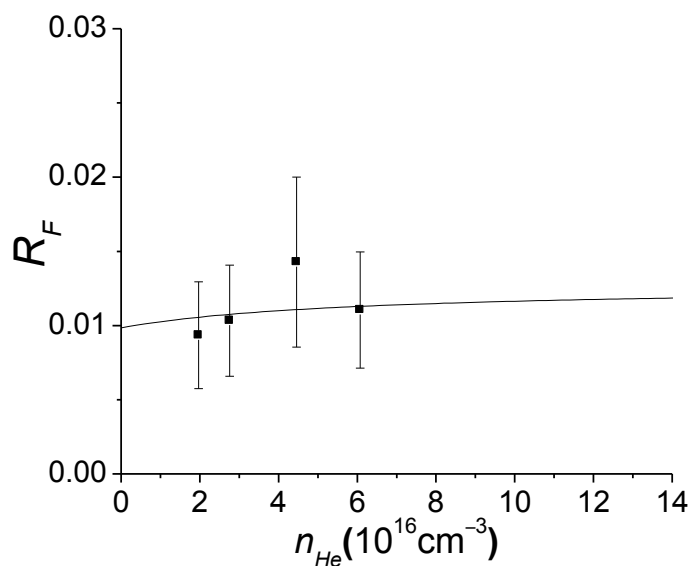
(a)  $n_K = 1.80 \times 10^{15} \text{ cm}^{-3}$



(b)  $n_K = 2.75 \times 10^{15} \text{ cm}^{-3}$



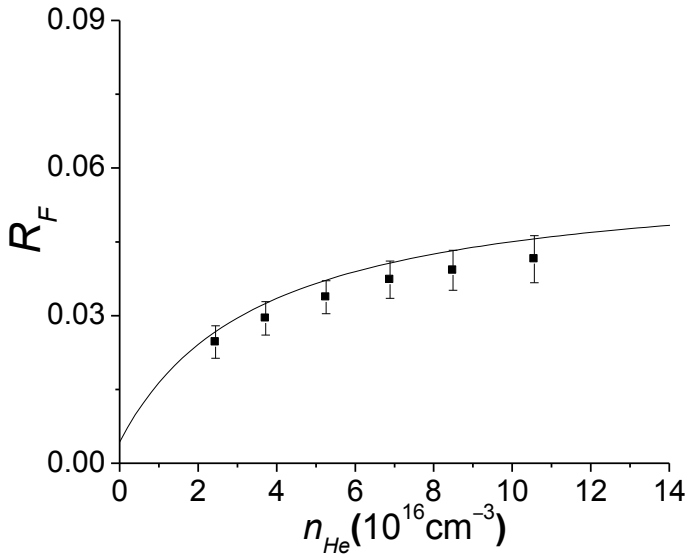
(c)  $n_K = 2.94 \times 10^{15} \text{ cm}^{-3}$



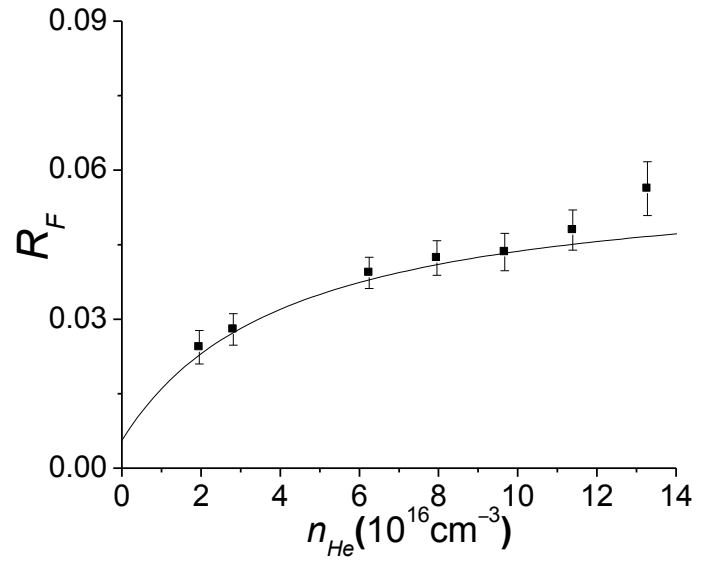
(d)  $n_K = 5.37 \times 10^{15} \text{ cm}^{-3}$

Supplementary Material Fig. 15: Plots of fluorescence ratio data ( $R_F$ ) versus helium density for  $\Delta J = -3$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ .

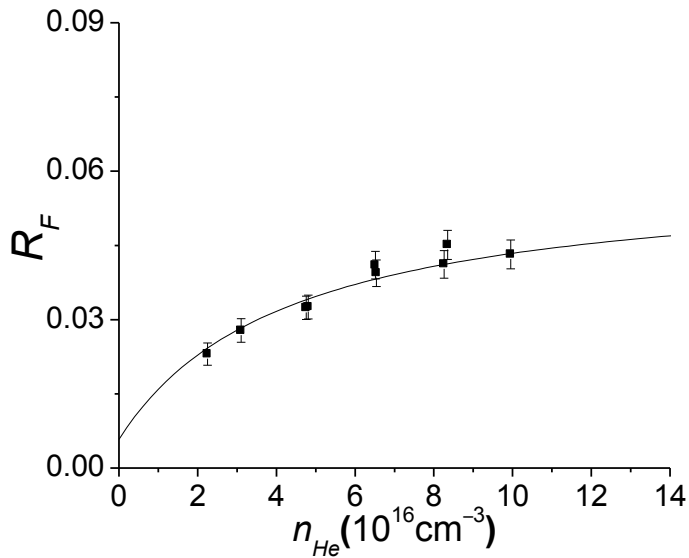
$R_F$  vs.  $n_{He}$  for NaK  $2(A)^1\Sigma^+(v=16, J=30)$ ,  $\Delta J = -4$



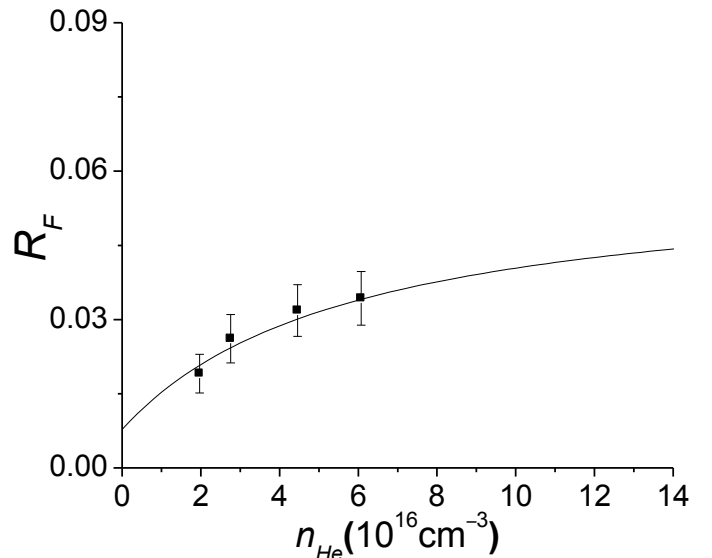
(a)  $n_K = 1.80 \times 10^{15} \text{ cm}^{-3}$



(b)  $n_K = 2.75 \times 10^{15} \text{ cm}^{-3}$



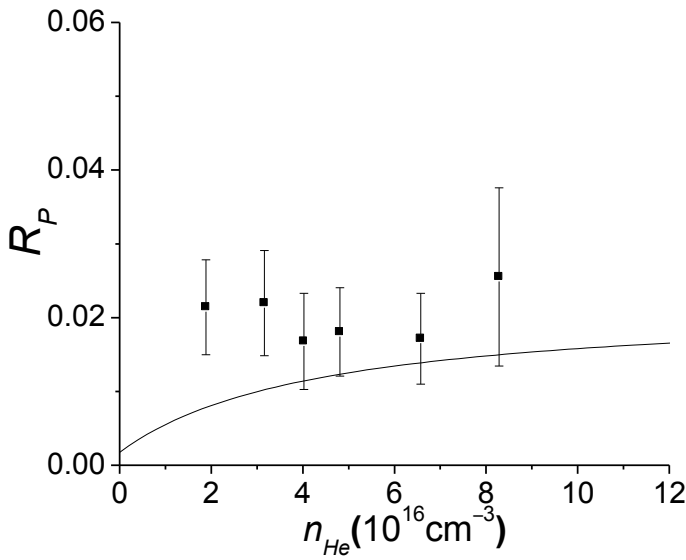
(c)  $n_K = 2.94 \times 10^{15} \text{ cm}^{-3}$



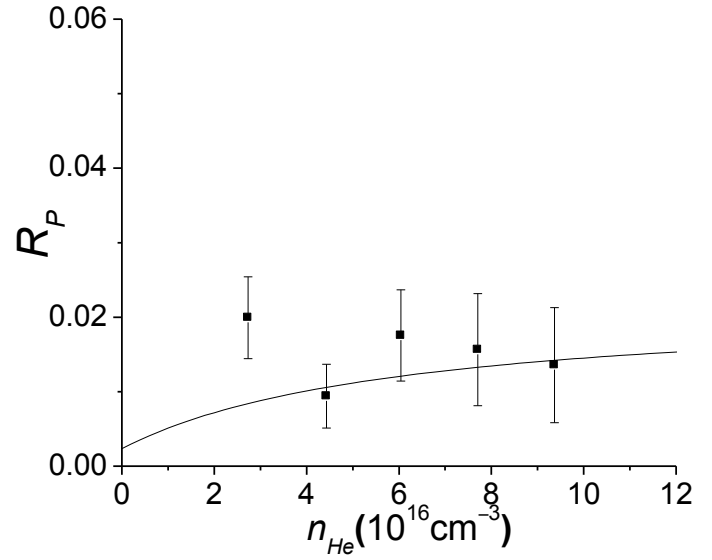
(d)  $n_K = 5.37 \times 10^{15} \text{ cm}^{-3}$

Supplementary Material Fig. 16: Plots of fluorescence ratio data ( $R_F$ ) versus helium density for  $\Delta J = -4$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ .

### $R_p$ vs. $n_{He}$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$ , $\Delta J = +1$



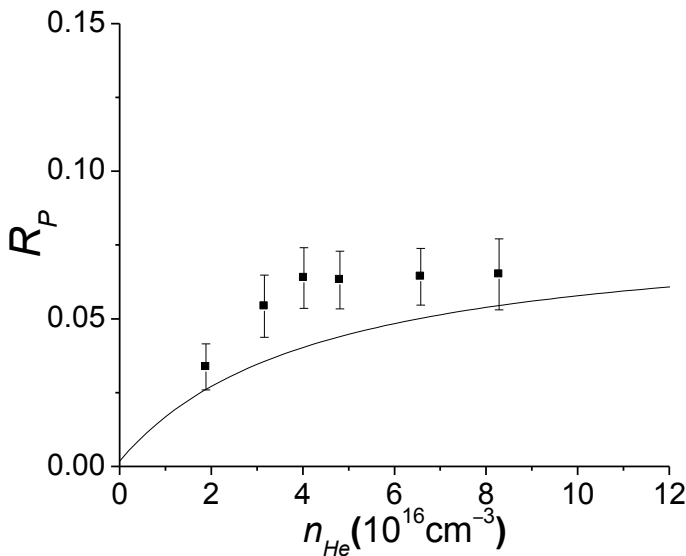
(a)  $n_K = 2.88 \times 10^{15} \text{ cm}^{-3}$



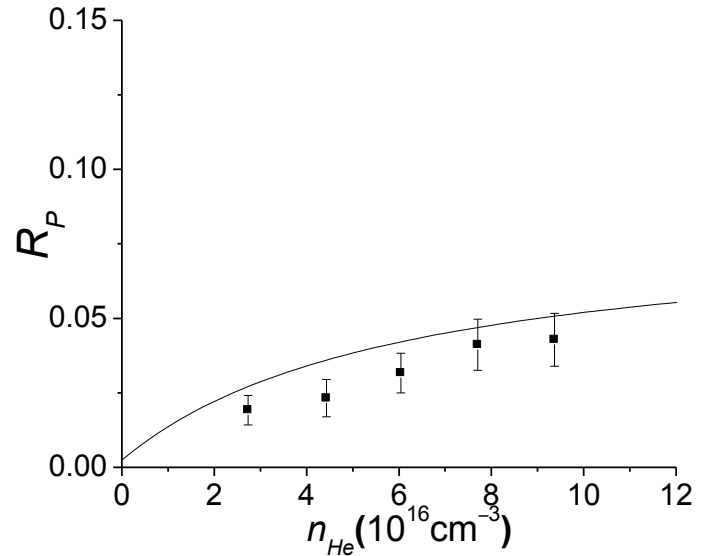
(b)  $n_K = 5.44 \times 10^{15} \text{ cm}^{-3}$

Supplementary Material Fig. 17: Plots of polarization spectroscopy ratio data ( $R_p$ ) versus helium density for  $\Delta J = +1$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ . In this, and subsequent plots, the calculated  $R_p$  values (solid curves) were obtained using the global fit parameters given in Tables 1 and 2 of the paper (also listed in Supplementary Tables 1 and 2).

### $R_p$ vs. $n_{He}$ for NaK $2(A)^1\Sigma^+(v=16, J=30)$ , $\Delta J = +2$



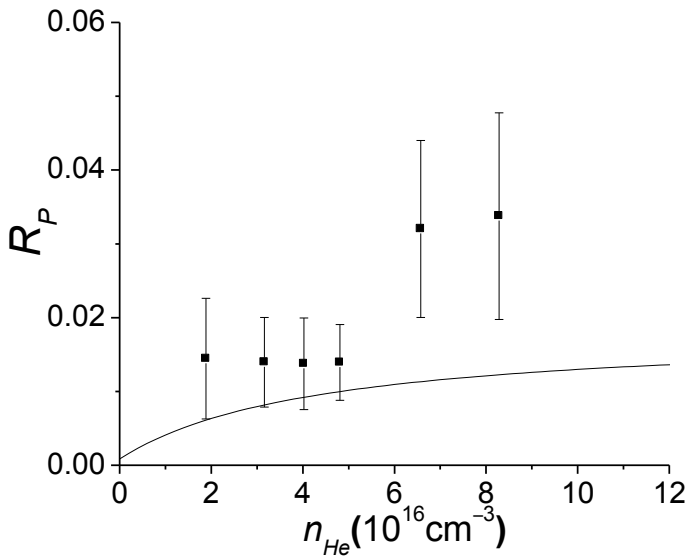
(a)  $n_K = 2.88 \times 10^{15} \text{ cm}^{-3}$



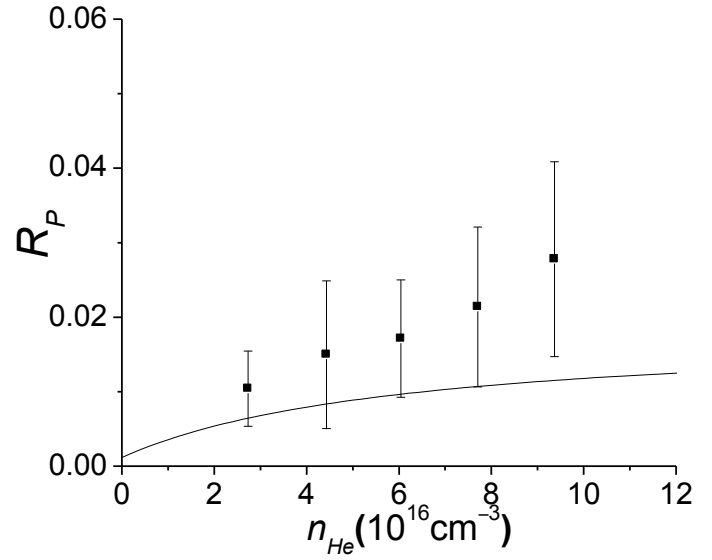
(b)  $n_K = 5.44 \times 10^{15} \text{ cm}^{-3}$

Supplementary Material Fig. 18: Plots of polarization spectroscopy ratio data ( $R_p$ ) versus helium density for  $\Delta J = +2$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ .

$R_P$  vs.  $n_{He}$  for NaK  $2(A)^1\Sigma^+(v=16, J=30)$ ,  $\Delta J = +3$



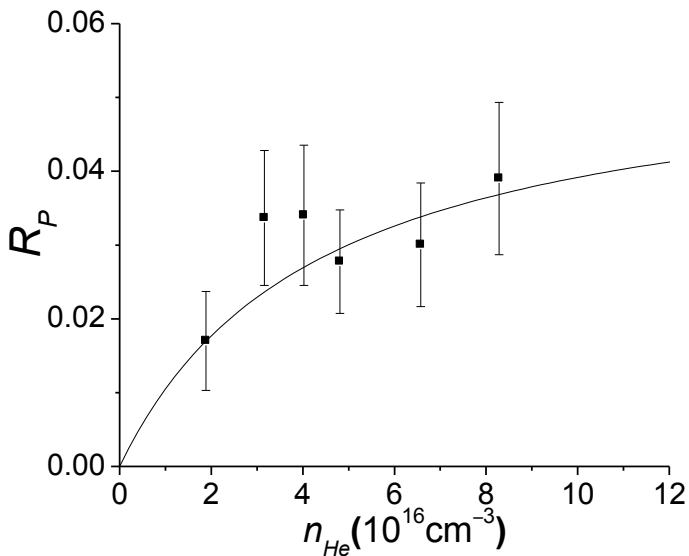
(a)  $n_K = 2.88 \times 10^{15} \text{ cm}^{-3}$



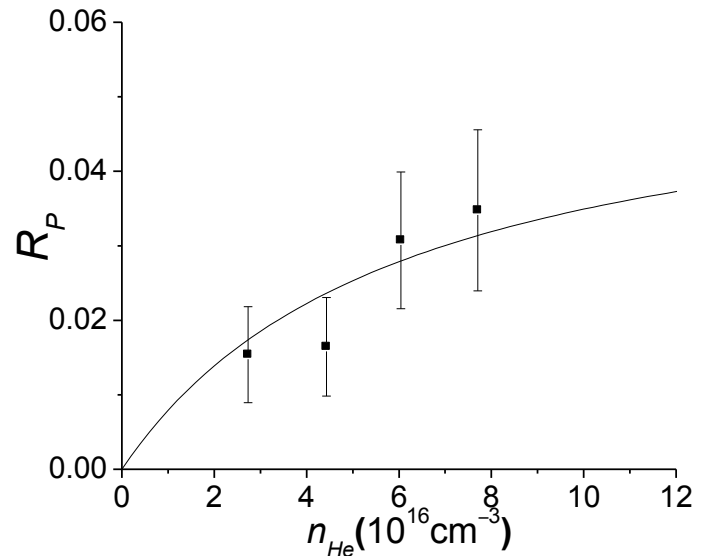
(b)  $n_K = 5.44 \times 10^{15} \text{ cm}^{-3}$

Supplementary Material Fig. 19: Plots of polarization spectroscopy ratio data ( $R_P$ ) versus helium density for  $\Delta J = +3$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ .

$R_P$  vs.  $n_{He}$  for NaK  $2(A)^1\Sigma^+(v=16, J=30)$ ,  $\Delta J = +4$



(a)  $n_K = 2.88 \times 10^{15} \text{ cm}^{-3}$

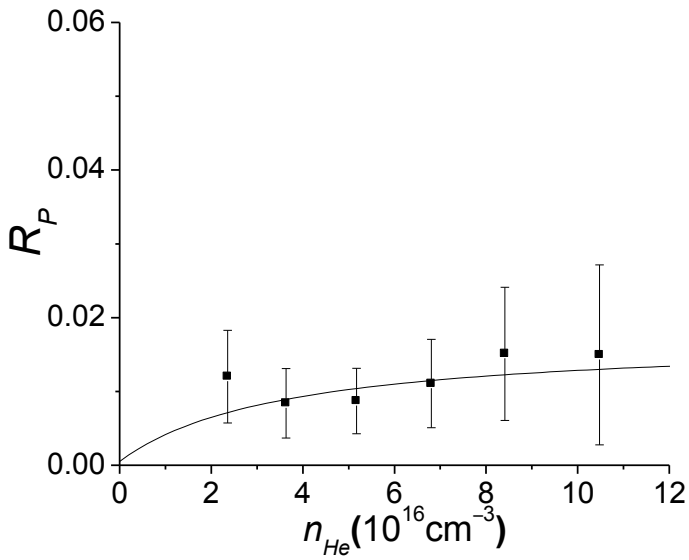


(b)  $n_K = 5.44 \times 10^{15} \text{ cm}^{-3}$

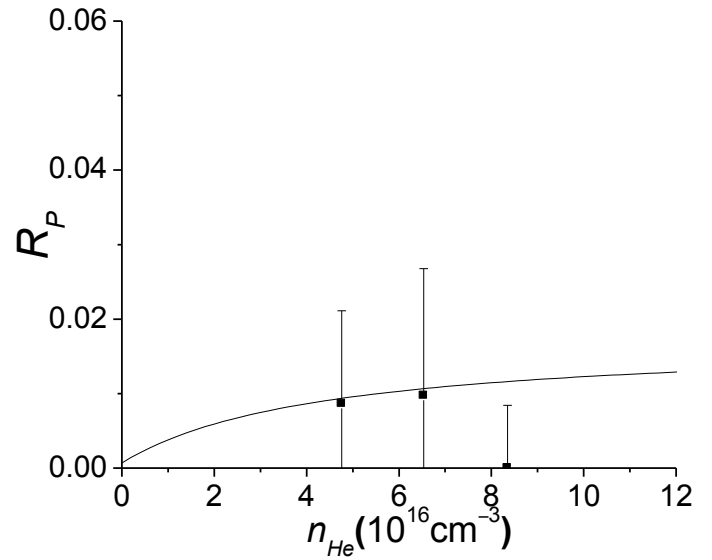
Supplementary Material Fig. 20: Plots of polarization spectroscopy ratio data ( $R_P$ ) versus helium density for  $\Delta J = +4$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ .



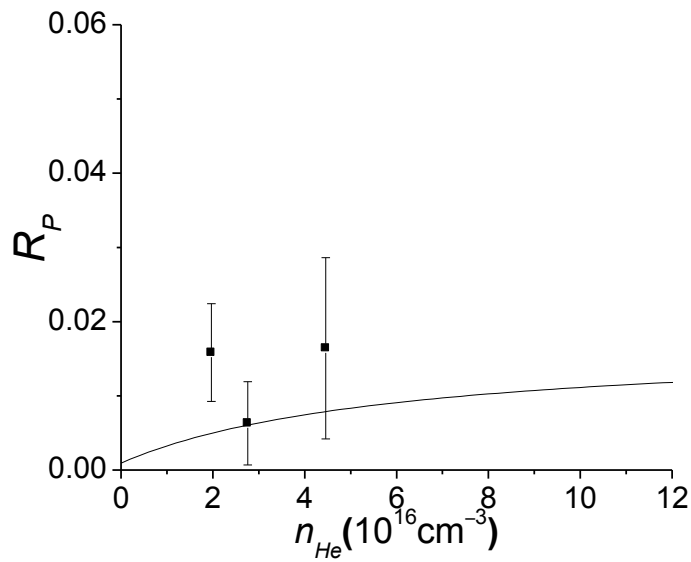
$R_p$  vs.  $n_{He}$  for NaK  $2(A)^1\Sigma^+(v=16, J=30)$ ,  $\Delta J = -1$



(a)  $n_K = 1.80 \times 10^{15} \text{ cm}^{-3}$



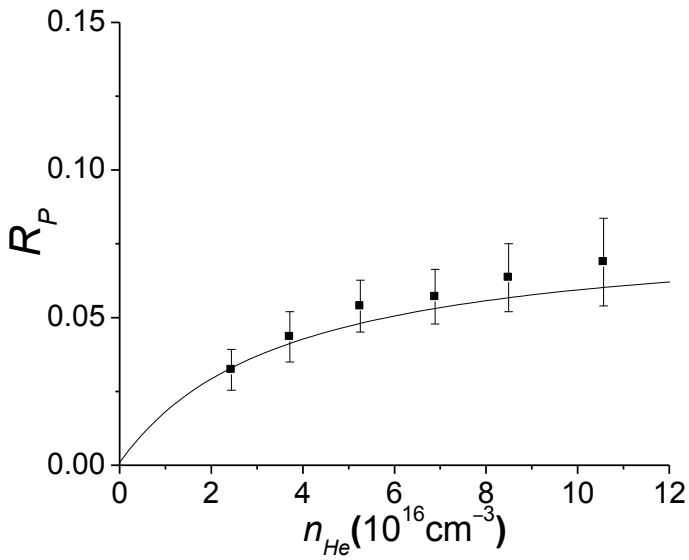
(b)  $n_K = 2.88 \times 10^{15} \text{ cm}^{-3}$



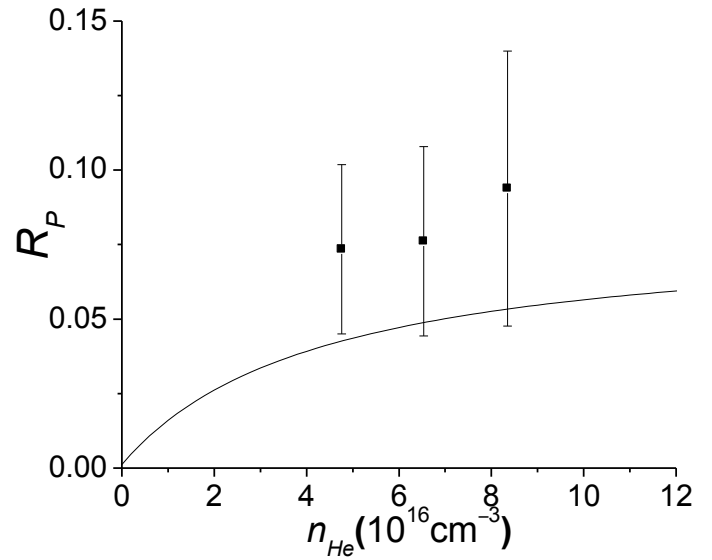
(c)  $n_K = 5.37 \times 10^{15} \text{ cm}^{-3}$

Supplementary Material Fig. 21: Plots of polarization spectroscopy ratio data ( $R_p$ ) versus helium density for  $\Delta J = -1$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ .

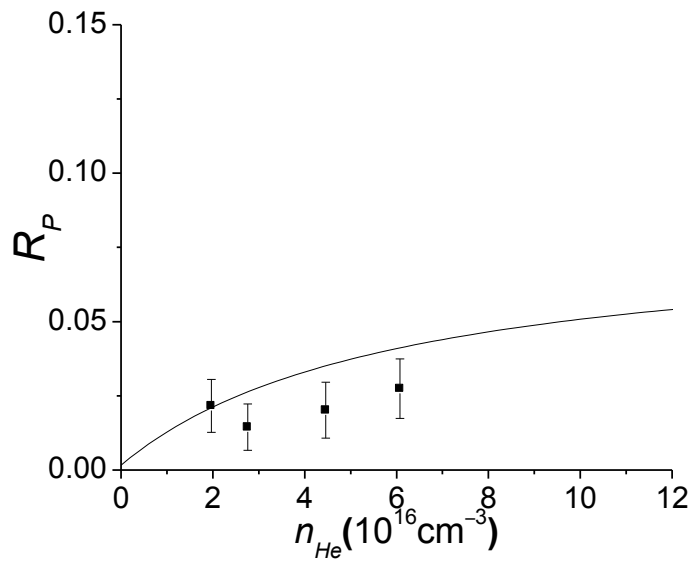
$R_P$  vs.  $n_{He}$  for NaK  $2(A)^1\Sigma^+(v=16, J=30)$ ,  $\Delta J = -2$



(a)  $n_K = 1.80 \times 10^{15} \text{ cm}^{-3}$



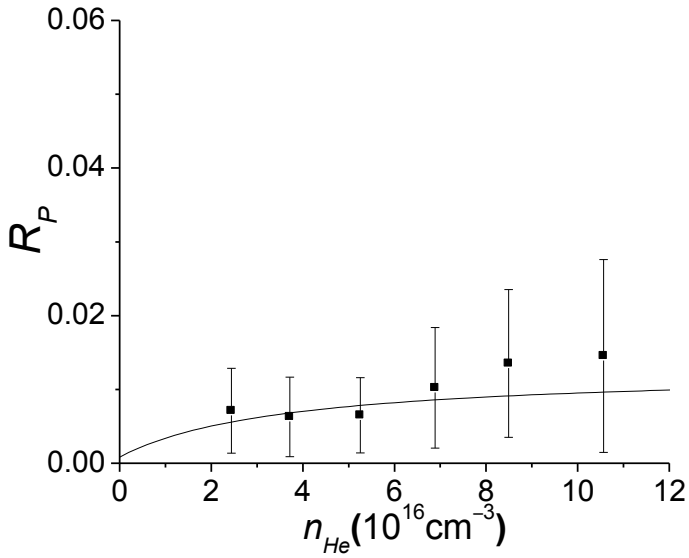
(b)  $n_K = 2.88 \times 10^{15} \text{ cm}^{-3}$



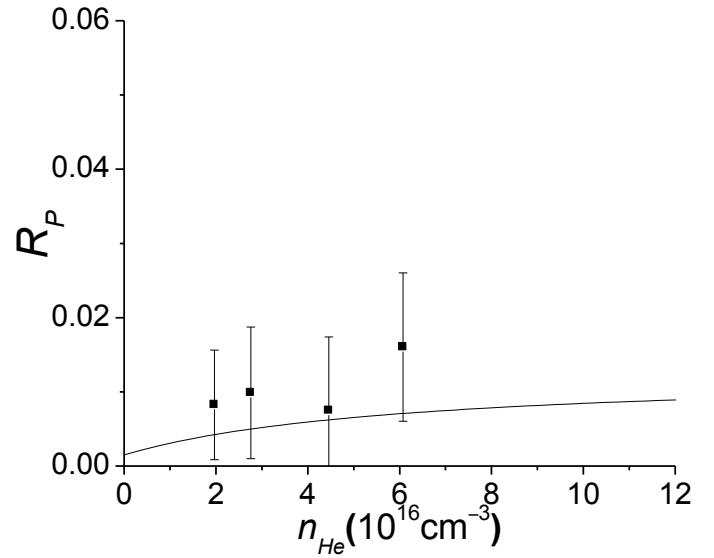
(c)  $n_K = 5.37 \times 10^{15} \text{ cm}^{-3}$

Supplementary Material Fig. 22: Plots of polarization spectroscopy ratio data ( $R_P$ ) versus helium density for  $\Delta J = -2$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ .

$R_p$  vs.  $n_{He}$  for NaK  $2(A)^1\Sigma^+(v=16, J=30)$ ,  $\Delta J = -3$



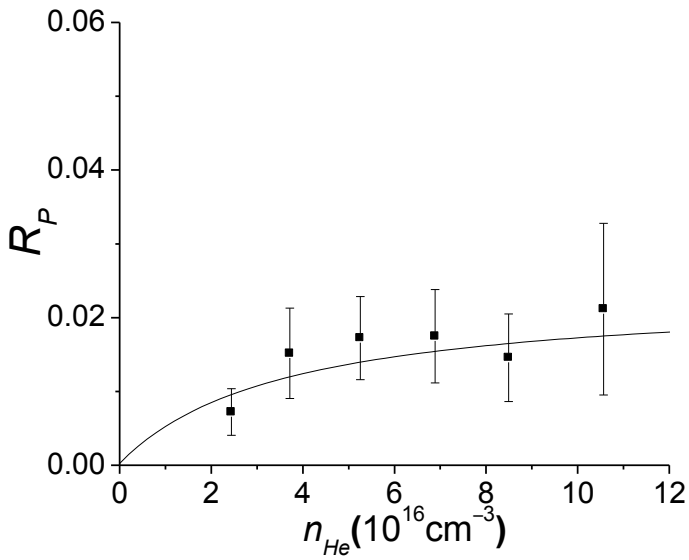
(a)  $n_K = 1.80 \times 10^{15} \text{ cm}^{-3}$



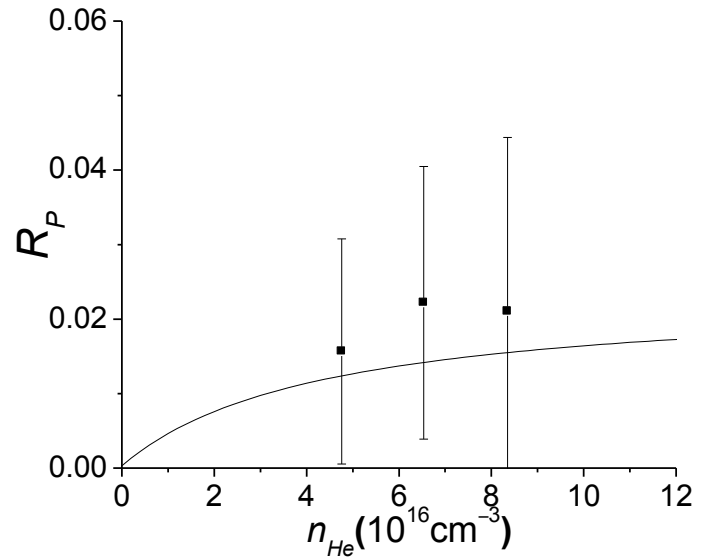
(b)  $n_K = 5.37 \times 10^{15} \text{ cm}^{-3}$

Supplementary Material Fig. 23: Plots of polarization spectroscopy ratio data ( $R_p$ ) versus helium density for  $\Delta J = -3$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ .

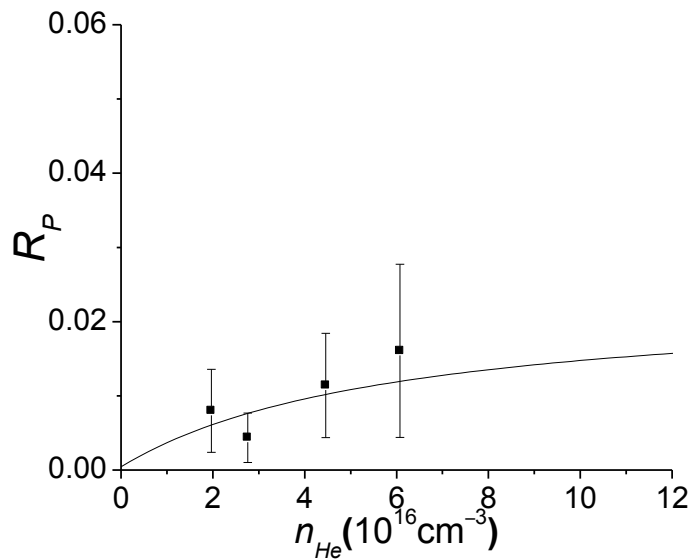
$R_P$  vs.  $n_{He}$  for NaK  $2(A)^1\Sigma^+(v=16, J=30)$ ,  $\Delta J = -4$



(a)  $n_K = 1.80 \times 10^{15} \text{ cm}^{-3}$

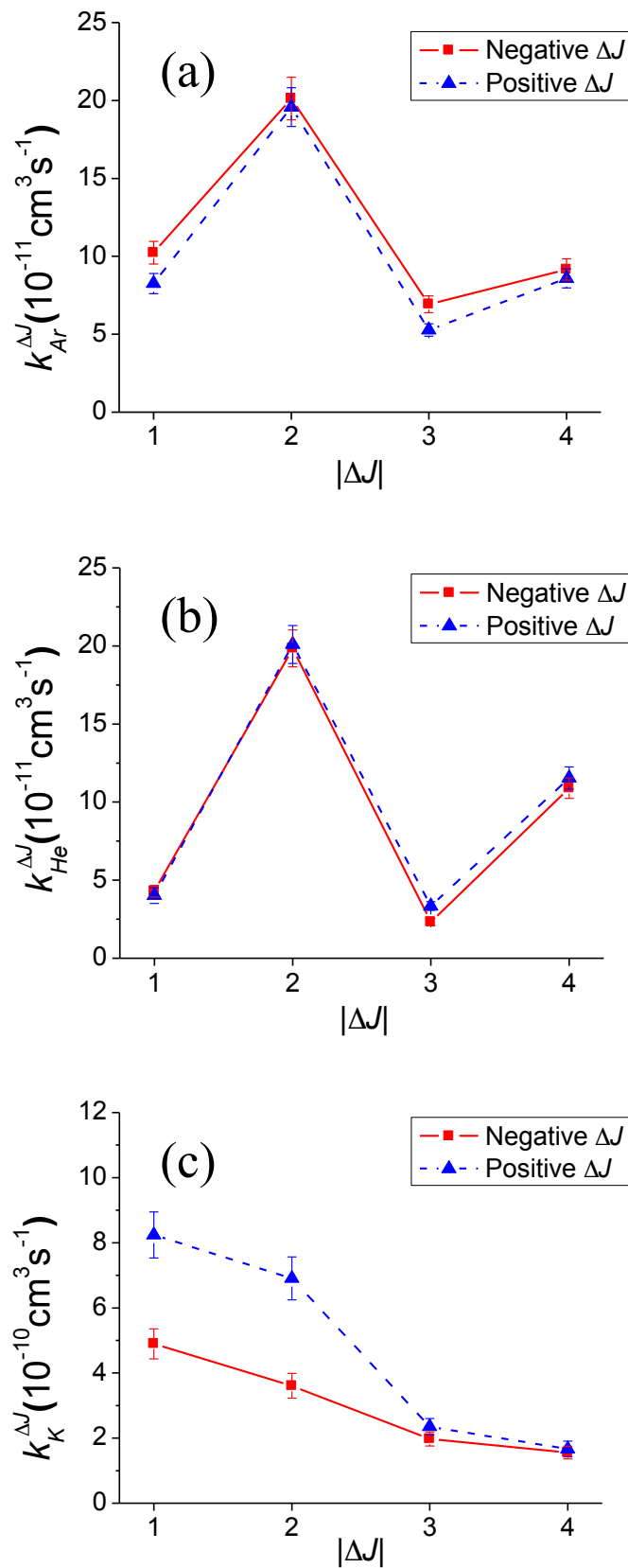


(b)  $n_K = 2.88 \times 10^{15} \text{ cm}^{-3}$

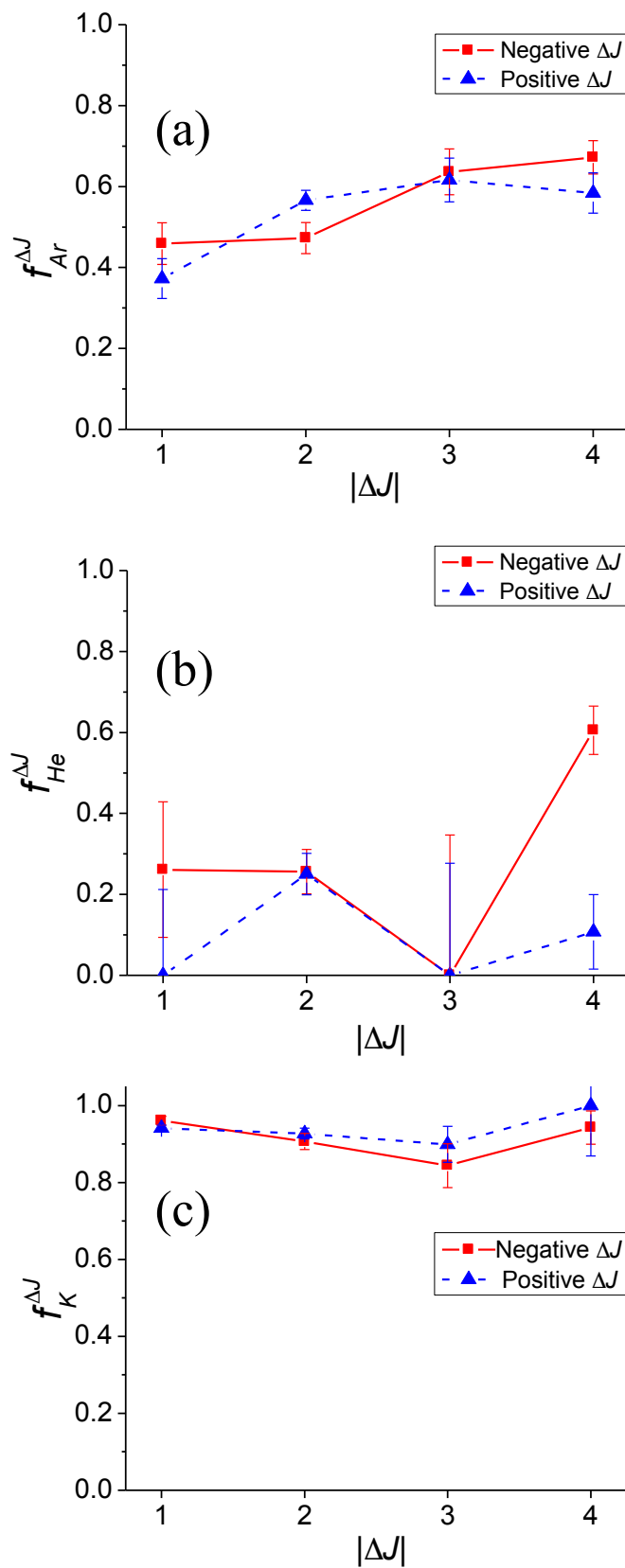


(c)  $n_K = 5.37 \times 10^{15} \text{ cm}^{-3}$

Supplementary Material Fig. 24: Plots of polarization spectroscopy ratio data ( $R_P$ ) versus helium density for  $\Delta J = -4$  collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with helium and potassium perturbers. Each panel represents a fixed potassium density  $n_K$ .



Supplementary Material Fig. 25: Experimental rate coefficients,  $k_{Ar}^{\Delta J}$ ,  $k_{He}^{\Delta J}$ , and  $k_K^{\Delta J}$ , for collisions of NaK  $2(A)^1\Sigma^+(v=16, J=30)$  molecules with (a) argon, (b) helium, and (c) potassium atoms as functions of  $\Delta J$ .



Supplementary Material Fig. 26: Experimental values for the fraction of orientation destroyed,  $f_{Ar}^{\Delta J}$ ,  $f_{He}^{\Delta J}$ , and  $f_K^{\Delta J}$ , in rotationally inelastic collisions of NaK  $2(A)1\Sigma^+(v=16, J=30)$  molecules with (a) argon, (b) helium, and (c) potassium atoms as functions of  $\Delta J$ .