Supplementary Materials: Ground and first excited states of the NaSr molecules. Experimental and theoretical study

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I. THE COMPOSITE COUPLED-CLUSTER (CCC) CALCULATIONS

TABLE I. The interaction energies at interatomic separation equal 7.5 bohr (approx. 4 Å) and the excitation energy of Sr predicted by different methods and basis sets. The atomic excitation energies are calculated in the dimer basis set.

Na	Sr	mb	SCF	$X^2\Sigma$		$2^2\Sigma^+$	^3P
				CCSD(T)	CCSD	EOM-CCSD	EOM-CCSD
aug-cc-pwCVTZ	aug-cc-pwCVTZ-PP	-	UHF	-1401.0	-1255.4	-5698.6	14546.7
aug-cc-pwCVQZ	aug-cc-pwCVQZ-PP	-	UHF	-1465.5	-1306.7	-5749.7	14595.6
aug-cc-pwCV5Z	aug-cc-pwCV5Z-PP	-	UHF	-1494.70	-1332.6	-5762.5	14582.5
aug-cc-pCVTZ	aug-cc-pCVTZ-PP	-	UHF	-1411.5	-1267.6	-5689.9	14444.5
aug-cc-pCVQZ	aug-cc-pCVQZ-PP	-	UHF	-1474.5	-1317.1	-5731.0	14536.8
aug-cc-pCV5Z	aug-cc-pCV5Z-PP	-	UHF	-1496.3	-1335.7	-5758.5	14561.7
aug-cc-pCVTZ	aug-cc-pwCVTZ-PP	-	UHF	-1397.2	-1257.1	-5736.3	14546.6
aug-cc-pCVQZ	aug-cc-pwCVQZ-PP	-	UHF	-1465.5	-1306.7	-5749.7	14595.6
aug-cc-pCV5Z	aug-cc-pwCV5Z-PP	-	UHF	-1492.9	-1331.7	-5765.0	14582.5
aug-pCV6Z	aug-pwCV6Z-PP	-	UHF	-1504.0	-1341.5	**-5771.2**	14579.6
aug-cc-pwCVQZ	aug-cc-pwCVQZ-PP	mb3	UHF		-1318.8	-5762.2	14595.8
aug-cc-pwCV5Z	aug-cc-pwCV5Z-PP	mb3	UHF		-1335.5	-5766.2	14583.1
aug-cc-pwCVQZ	aug-cc-pwCVQZ-PP	mb2	UHF		-1326.9	-5769.6	14595.3
aug-cc-pwCV5Z	aug-cc-pwCV5Z-PP	mb2	UHF		-1339.0	-5769.5	14582.8
aug-pCV6Z	aug-pwCV6Z-PP	mb2	UHF		-1342.0		14579.7
aug-cc-pwCVTZ	aug-cc-pwCVTZ-PP	-	ROHF	-1426.2	-1170.1		
aug-cc-pwCVQZ	aug-cc-pwCVQZ-PP	-	ROHF	-1494.1	-1220.4		
aug-cc-pwCV5Z	aug-cc-pwCV5Z-PP	-	ROHF	-1523.7	-1244.0		
aug-cc-pCVTZ	aug-cc-pwCVTZ-PP	-	ROHF	-1420.7	-1169.1		
aug-cc-pCVQZ	aug-cc-pwCVQZ-PP	-	ROHF	-1490.1	-1218.6		
aug-cc-pCV5Z	aug-cc-pwCV5Z-PP	-	ROHF	-1521.8	-1242.8		
aug-pCV6Z	aug-pwCV6Z-PP	-	ROHF	-1533.5	-1251.9		
aug-cc-pCV5Z	aug-cc-pwCV5Z-PP	mb3	ROHF	-1525.7	-1246.0		
aug-pCV6Z	aug-pwCV6Z-PP	mb3	ROHF	-1534.4	-1252.5		
aug-cc-pCV5Z	aug-cc-pwCV5Z-PP	mb2	ROHF	-1529.6	-1249.2		
aug-pCV6Z	aug-pwCV6Z-PP	mb2	ROHF	**-1536.5**	-1254.3		

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TABLE II. The CCSDT interaction energies of $X^2\Sigma^+$ state (cm^{-1}).

Na	Sr	SCF	$X^2\Sigma$
aug-cc-pwCVTZ	aug-cc-pwCVTZ-PP	UHF	-1501.5
aug-cc-pCVTZ	aug-cc-pCVTZ-PP	UHF	-1510.0
aug-cc-pCVTZ	aug-cc-pwCVTZ-PP	UHF	-1496.8
aug-cc-pwCVTZ	aug-cc-pwCVTZ-PP	ROHF	-1492.3
aug-cc-pCVTZ	aug-cc-pCVTZ-PP	ROHF	-1499.9
aug-cc-pCVTZ	aug-cc-pwCVTZ-PP	ROHF	-1487.5
aug-cc-pwCVQZ	aug-cc-pwCVQZ-PP	ROHF	-1552.5

TABLE III. The exponents of mid-bond functions.

			mb2		
s	0.0327	0.0749	0.1716	0.3930	0.9001
p	0.0496	0.1303	0.3425	0.9000	
d	0.0546	0.1810	0.6000		
f	0.3				
			mb3		
s	0.03757	0.0779	0.1457		
p	0.05173	0.1793	0.2568		
d	0.0671	0.1482			

II. THE (ECP+CPP)FCI CALCULATIONS

TABLE IV. The Na+Sr asymptotic energies.

Na	Sr	E [Hartre]
$3s\ ^2S$	$5s^2\ ^1S$	-0.8041347
$3s\ ^2S$	$5s5p\ ^3P$	-0,7376522
$3p\ ^2P$	$5s^2\ ^1S$	-0,7268213
$3s\ ^2S$	$5s4d\ ^3D$	-0,7186226
$3s\ ^2S$	$5s4d\ ^1D$	-0,7094944
$3s\ ^2S$	$5s5p\ ^1P$	-0,7065121

III. SIMULATIONS

[1] J. V. Pototschnig, R. Meyer, A. W. Hauser, and W. E. Ernst, Vibronic transitions in the alkali-metal (Li, Na, K, Rb) ˘ alkaline-earth-metal (Ca, Sr) series: A systematic analysis of de-excitation mechanisms based on the graphical mapping of Frank-Condon integrals, Phys. Rev. A **95**, 022501 (2017).

[2] I. Zeid, T. Atallah, S. Kontar, W. Chmaisani, N. El-Kork, and M. Korek, Theoretical electronic structure of the molecules SrX (X= Li, Na, K) toward laser cooling study, Comput. Theor. Chem. **1126**, 16 (2018).

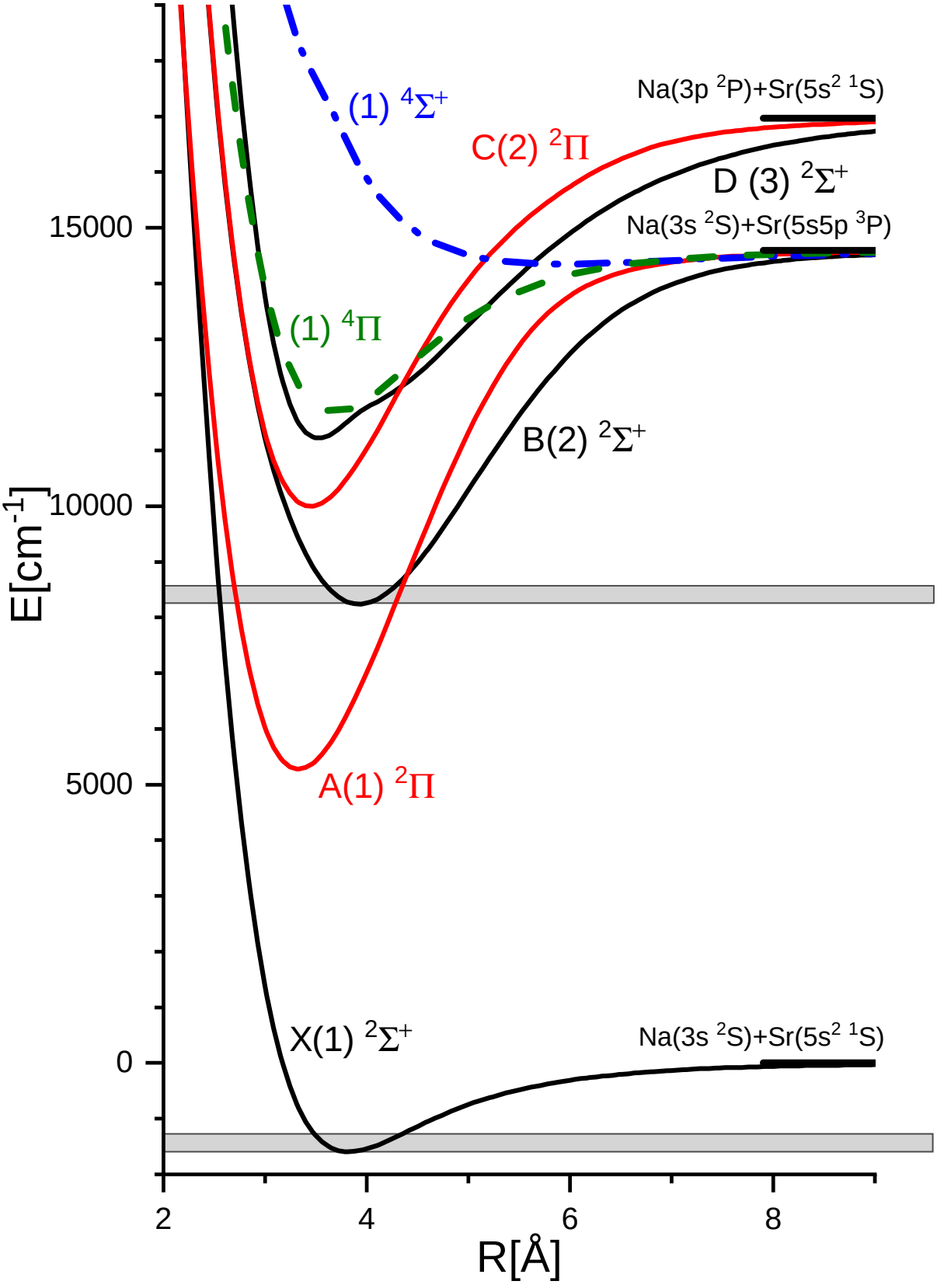

FIG. 1. PECs of the ground state and excited states of NaSr up to the Na(3p ^2P)+Sr(5s^2 ^1P) dissociation limit calculated with the (ECP+CPP)FCI method. The grey-shaded area corresponds to the energy range studied experimentally.

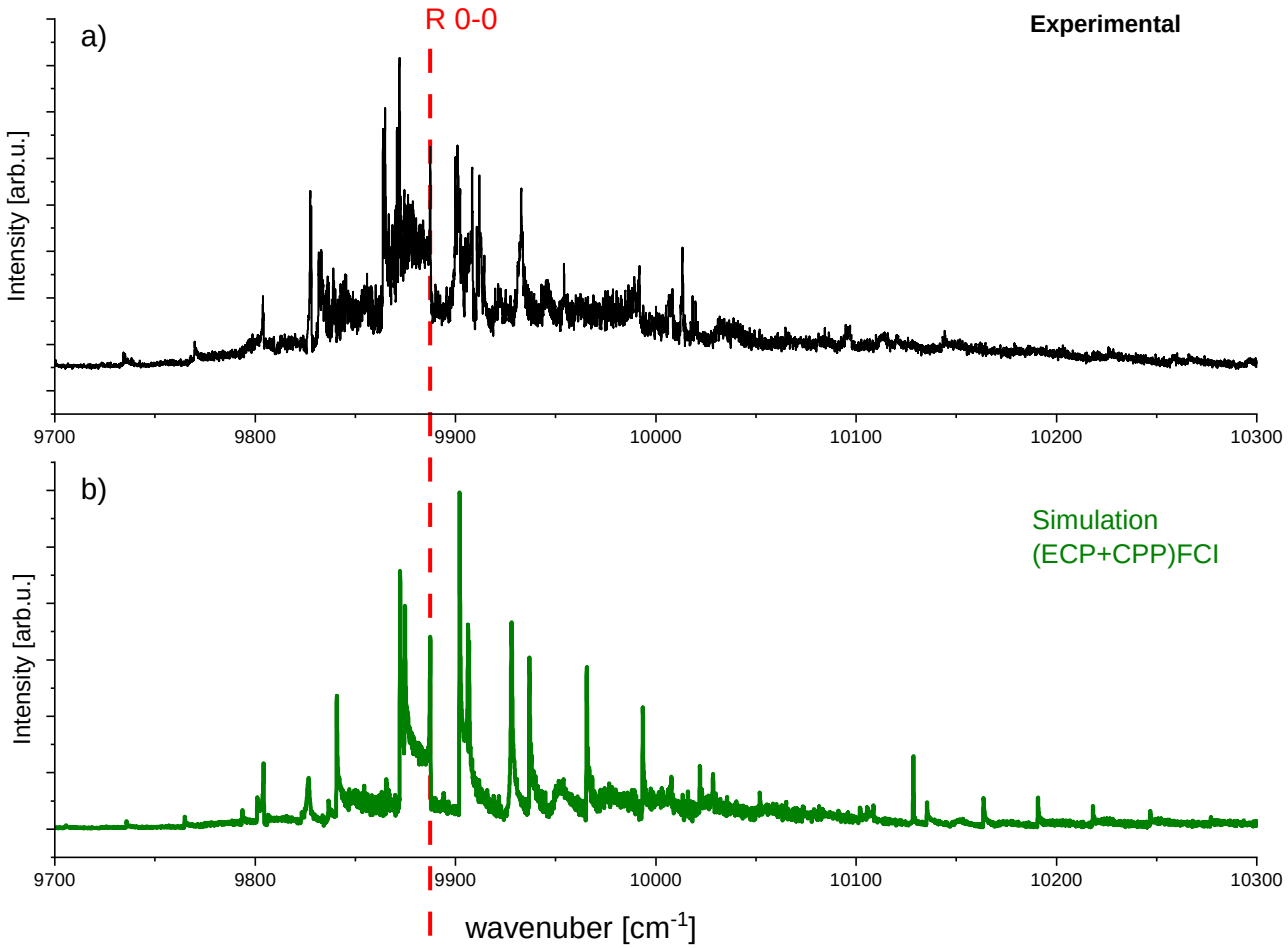

FIG. 2. Comparison of the experimental thermoluminescence spectrum (a) with simulated spectra based on theoretical curves. The theoretical spectra were shifted so that the onset of the 0-0 band coincides with its the position in the experimental spectrum: (b) spectrum resulting from the (ECP+CPP)FCI calculations (this work) shifted by $\approx +26\,\mathrm{cm}^{-1}$

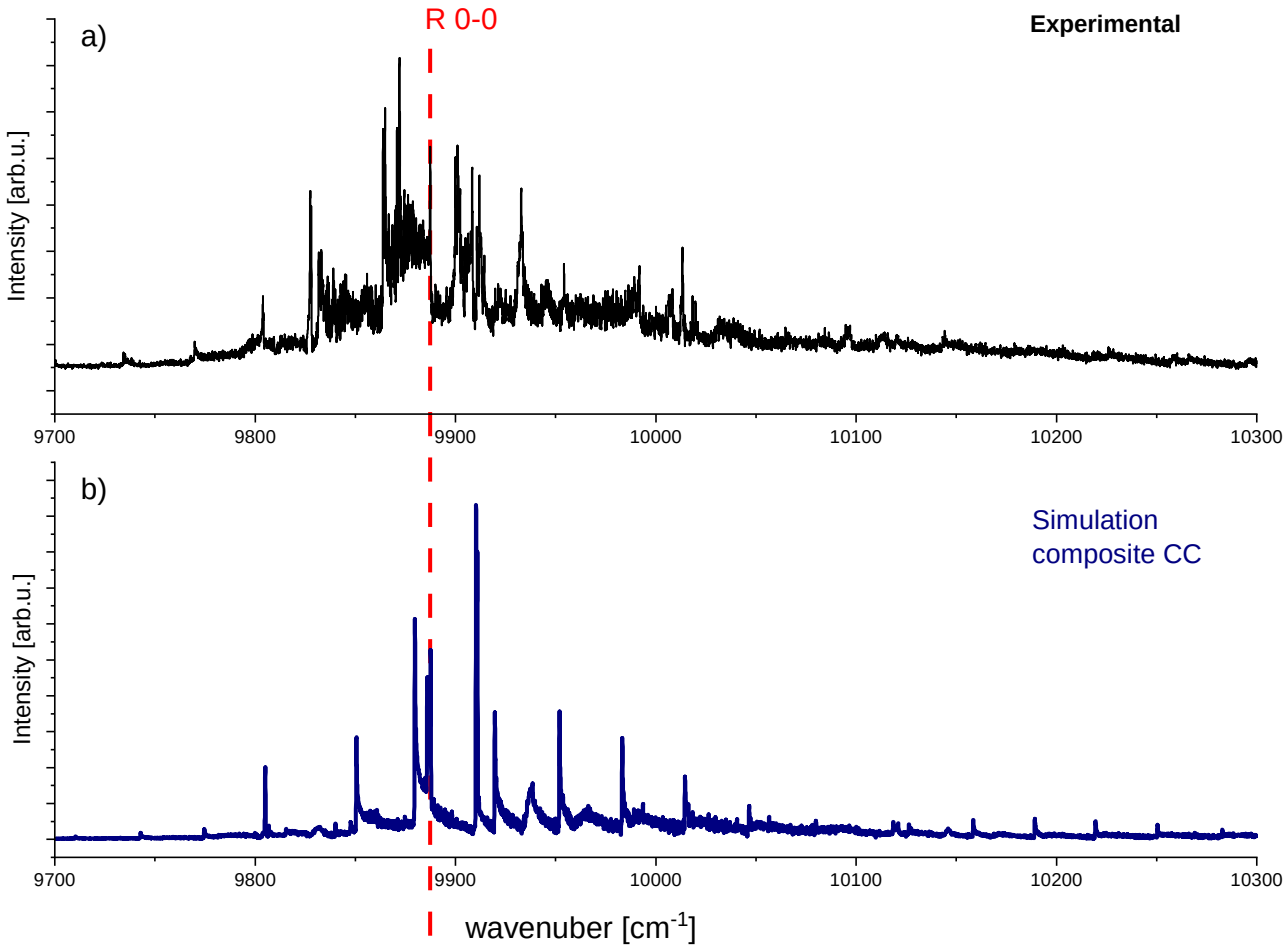

FIG. 3. Comparison of the experimental thermoluminescence spectrum (a) with simulated spectra based on theoretical curves. The theoretical spectra were shifted so that the onset of the 0-0 band coincides with its the position in the experimental spectrum: (b) spectrum resulting from the composite CC calculations (this work) shifted by $\approx -251\,\mathrm{cm}^{-1}$

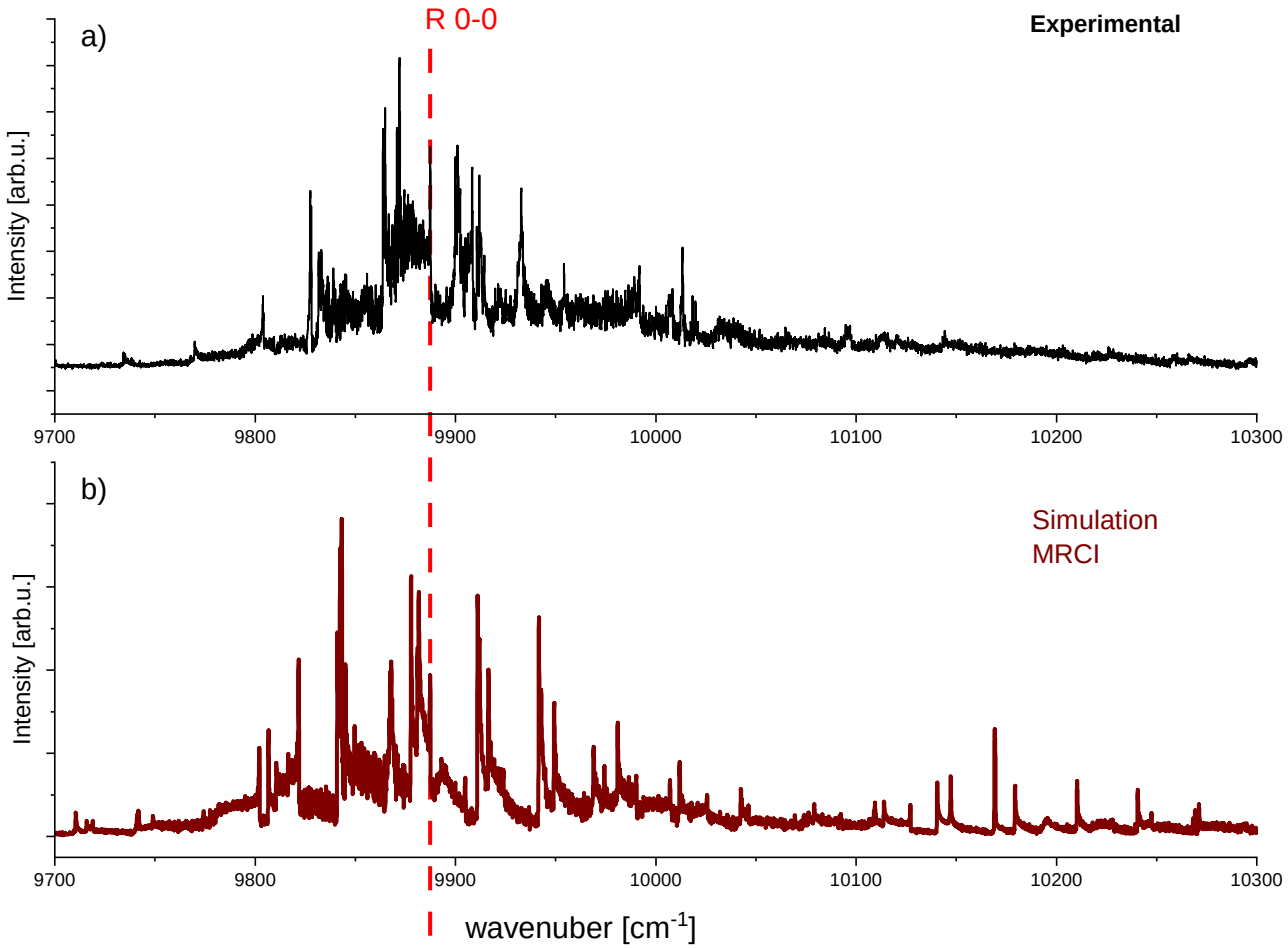

FIG. 4. Comparison of the experimental thermoluminescence spectrum (a) with simulated spectra based on theoretical curves. The theoretical spectra were shifted so that the onset of the 0-0 band coincides with its the position in the experimental spectrum: (b) spectrum resulting from the MRCI [1] shifted by $\approx +71\,\mathrm{cm}^{-1}$

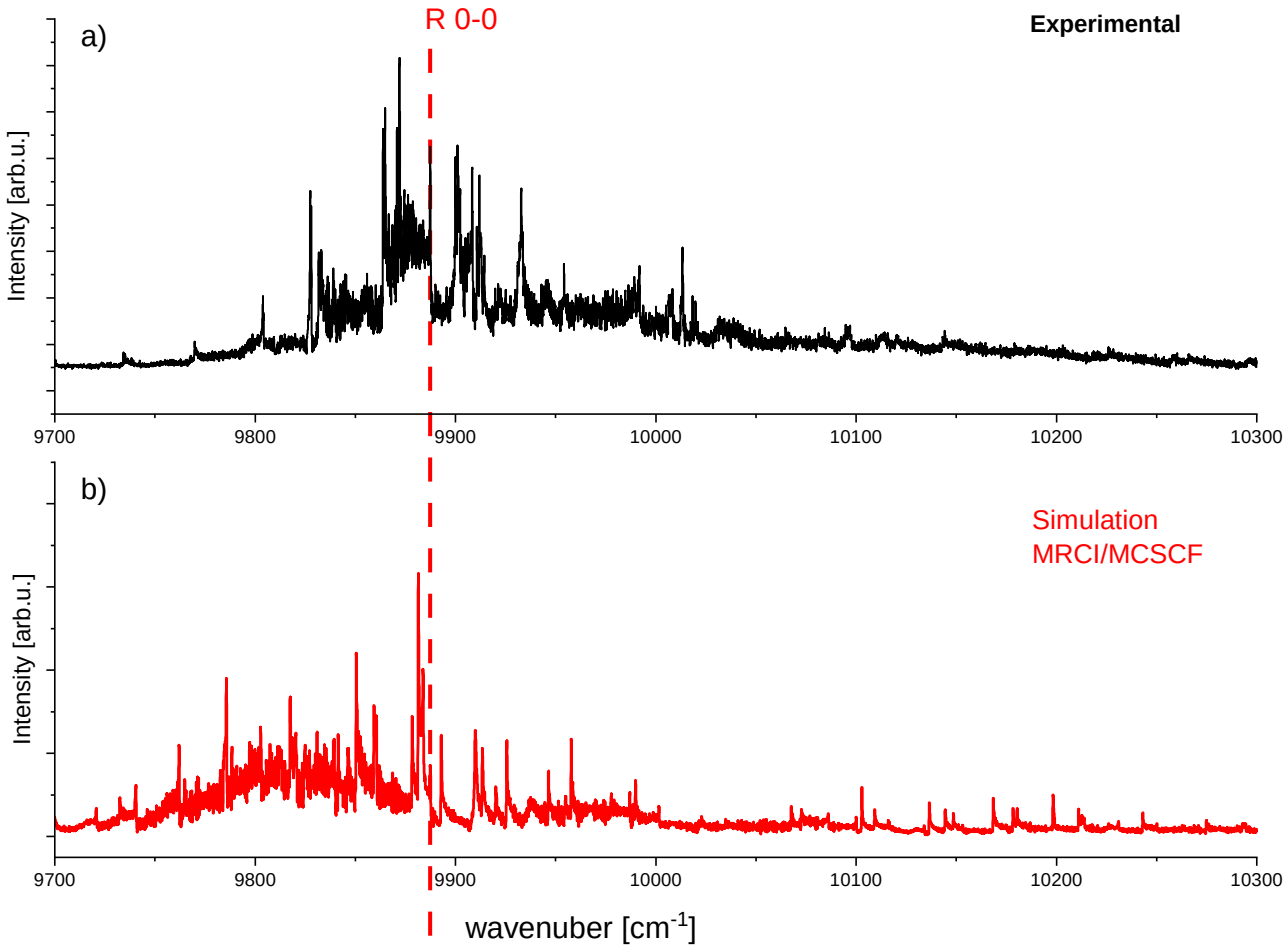

FIG. 5. Comparison of the experimental thermoluminescence spectrum (a) with simulated spectra based on theoretical curves. The theoretical spectra were shifted so that the onset of the 0-0 band coincides with its the position in the experimental spectrum: (b) spectrum resulting from the MRCI/MCSCF [2] shifted by $\approx -687\,\mathrm{cm}^{-1}$