## **Supporting Information**

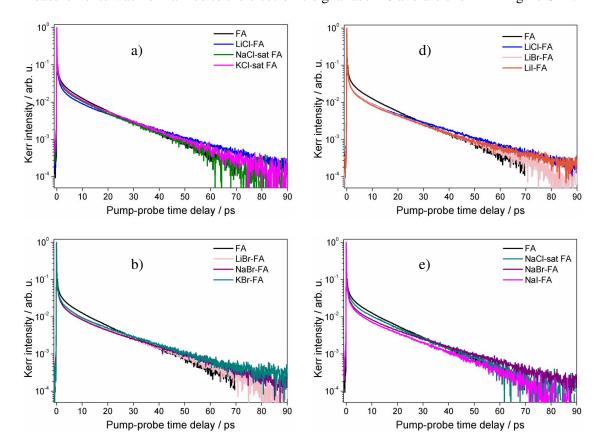
## Dynamics of formamide ionic solutions investigated by ultrafast optical Kerr effect

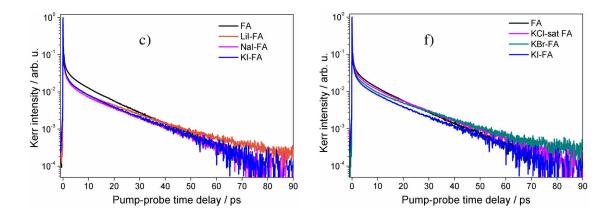
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**Time response** The time-domain response of FA ionic solutions from OHD-OKE measurements was normalized to the electronic signal at t = 0 and are shown in Figure SI-1.





**Figure SI-1.** Kerr transient of alkali metal halide–FA solutions at 1.3 M concentration (except for NaCl and KCl, which gave rise to saturated solutions with 0.69 and 1.0 M concentration, respectively). (a-c) Same anion, different cation; (d-f) same cation, different anion.

**Density measurements** The density of FA ionic solutions used for the OHD-OKE measurements was measured at room temperature.

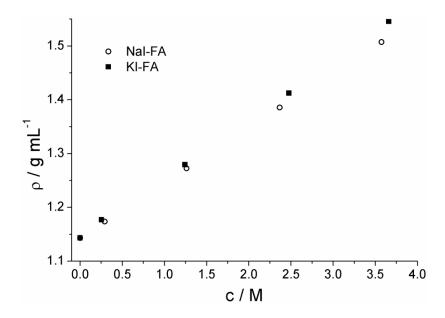
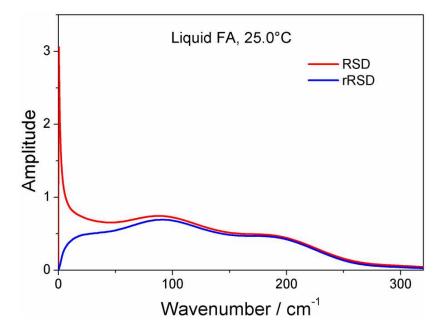
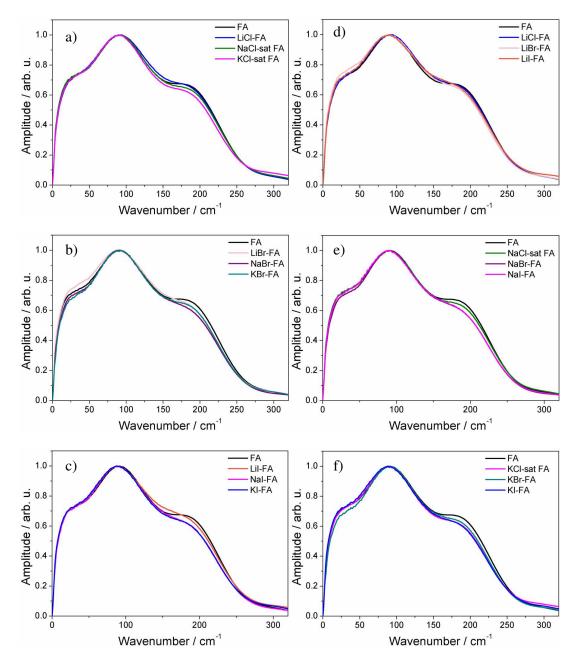


Figure SI-2. Plot of density of NaI and KI–FA solutions versus concentration. Error bars are within the symbol size.

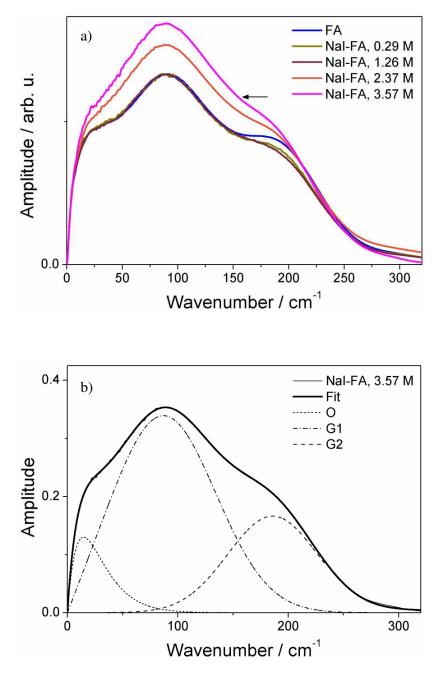
**Spectral response** The spectral response of FA ionic solutions derived from OHD-OKE measurements was investigated



*Figure SI-3. Full and reduced RSD of neat FA derived from the Kerr transient at 25.0°C.* 



*Figure SI-4.* Reduced RSD of alkali metal halide–FA solutions at 1.3 M concentration (except for NaCl and KCl, which gave rise to saturated solutions), and 25.0°C. Spectra were normalized to the maximum of the band at ~ 90 cm<sup>-1</sup>, which shows only minor changes between different samples. (a-c) Same anion, different cation; (d-f) same cation, different anion.



**Figure SI-5.** (a) Evolution of the rRSD of the NaI–FA solution as a function of concentration. Spectra were normalized to the intensity at lower frequencies, where only slight changes are observed. Arrow indicates the direction of the observed shift with increasing NaI concentration. (b) Results of a multicomponent fit applied to rRSD of a concentrated NaI–FA solution using a sum of an Ohmic and two anti-symmetrized Gaussians.

	<i>c /</i> M	$A_{\mathrm{O}}$	$v_0$	$A_{\rm G1}$	$v_{G1}$	$\Delta v_{G1}$	$A_{ m G2}$	$v_{G2}$	$\Delta v_{G2}$
			$/ \text{ cm}^{-1}$		$/ \text{ cm}^{-1}$	$/ \text{ cm}^{-1}$	-	$/ \text{ cm}^{-1}$	$/ \text{ cm}^{-1}$
FA		0.121	15.7	0.563	90.6	101.6	0.316	190.9	90.1
LiCl	1.33	0.109	15.5	0.600	91.0	108.8	0.292	192.2	88.9
NaCl	0.69	0.122	15.7	0.571	90.6	102.3	0.307	190.7	91.0
KCl	1.00	0.124	16.0	0.575	89.7	100.5	0.301	188.4	89.7
LiBr	1.28	0.113	15.1	0.600	89.1	109.6	0.287	190.1	90.6
NaBr	1.28	0.135	17.0	0.519	89.1	93.2	0.346	184.0	98.5
KBr	1.27	0.130	17.3	0.538	90.5	95.3	0.332	186.8	93.2
LiI	1.27	0.114	15.6	0.551	87.6	102.3	0.335	186.3	96.7
NaI	0.29	0.115	14.9	0.588	89.0	104.7	0.297	189.9	90.4
"	1.26	0.101	14.2	0.614	88.2	110.3	0.285	190.7	91.9
"	2.37	0.091	14.7	0.622	87.1	111.6	0.287	187.8	93.7
"	3.57	0.086	14.6	0.631	86.7	112.2	0.283	185.8	95.7
KI	0.25	0.124	16.0	0.555	90.1	99.7	0.321	189.3	91.8
"	1.24	0.127	16.1	0.560	88.6	99.7	0.313	186.7	93.2
"	2.48	0.104	16.1	0.568	87.2	99.7	0.328	183.8	95.1
"	3.66	0.100	16.0	0.567	85.9	98.1	0.332	179.4	97.2
	T ∕°C	$A_{\rm O}$	$\nu_{O}$	<b>A</b>	$\nu_{G1}$	$\Delta v_{G1}$	$A_{ m G2}$	$\nu_{G2}$	$\Delta v_{G2}$
	17 C	A()	$/ \text{ cm}^{-1}$	$A_{\rm G1}$	$/ \text{ cm}^{-1}$	$/ \text{ cm}^{-1}$		$/ \text{ cm}^{-1}$	$/ \text{ cm}^{-1}$
FA									
	5.0	0.138	17.0	0.551	93.2	97.2	0.311	192.2	88.5
	15.0	0.127	16.4	0.567	92.4	100.3	0.305	191.8	86.8
	25.0	0.121	15.7	0.563	90.6	101.6	0.316	190.9	90.1
	35.0	0.118	15.6	0.569	89.4	102.5	0.313	189.4	90.6
	45.0	0.127	16.0	0.550	87.4	98.6	0.323	185.2	94.4
	55.0	0.125	16.2	0.543	86.5	98.2	0.331	183.5	95.5
	65.0	0.120	15.9	0.535	84.6	97.4	0.345	180.7	98.5

FA solutions, at 25.0°C, and (b) pure liquid FA, at different temperatures

Values are reported as A: relative amplitude  $(a_i/\Sigma a_n)$ , where  $a_i$  is the integrated amplitude in the range 0.03 to 400 cm<sup>-1</sup>); v: characteristic frequency;  $\Delta v$ : band width parameter. O, G1, and G2 subscripts refer to the Ohmic and the two anti-symmetrized Gaussians, respectively.