Supporting Online Material for

The isotope Soret effect in molecular liquids: a quantum effect at room temperatures

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Experimental details

All measurements of the Soret coefficients have been performed by means of the transient holographic grating technique of Thermal Diffusion Forced Rayleigh Scattering [Köhler(1993), Wittko and Köhler(2003)]. With this experimental method, a transient holographic temperature grating is written into the solution and, as a consequence of the Soret effect, a concentration grating develops. The diffraction efficiency of the resulting phase grating, which contains contributions from both the temperature and the concentration grating, is read out by Bragg diffraction of a readout laser beam. The writing wavelength is 532 nm (Coherent Verdi V5) and the readout wavelength 633 nm (HeNe). Since a slight optical absorption at the writing wavelength is needed, a tiny amount of an inert dye (quinizarin) is added to the mixtures. The absence of an unwanted dye contribution to the signal has been checked both for the pure substances and for the mixtures by variation of the dye concentration. For improved signal amplitude and quality, a heterodyne detection scheme has been employed, where the phase between the coherent reference wave and the diffracted beam is actively stabilized [Köhler and Rossmanith(1995)]. The diffracted heterodyne signal $\zeta_{het}(t)$ has been fitted with the equation

$$\zeta_{het}(t) = 1 - \mathrm{e}^{-t/\tau_{th}} - A(\tau - \tau_{th})^{-1}\left[\tau\left(1 - \mathrm{e}^{-t/\tau}\right) - \tau_{th}\left(1 - \mathrm{e}^{-t/\tau_{th}}\right)\right] , \qquad (1)$$

where τ and τ_{th} are the mass and temperature diffusion times. The amplitude factor

$$A = \left(\frac{\partial n}{\partial c}\right)_{p,T}\left(\frac{\partial n}{\partial T}\right)^{-1}_{p,c} S_T\, c_0(1 - c_0) \qquad (2)$$

contains the Soret coefficient S_T as a fit parameter. c_0 is the mean concentration (weight fractions) of the mixture.

The two contrast factors $(\partial n/\partial c)_{p,T}$ and $(\partial n/\partial T)_{p,c}$ have been measured separately with refractometers (Anton Paar Abbemat or ATG Abbe-Refraktometer B) and an interferometer, respectively.

Values of the Soret coefficients and the contrast factors

The measured values of the contrast factors and the Soret coefficients for the systems described are summarized in Tab. S1. x and c are the concentration of the first component in mole and weight fraction, respectively. All uncertainties are standard errors. The errors of S_T define a lower limit and are obtained by error propagation of the measured stationary diffraction amplitudes, the contrast factors, and the concentration. The true errors are somewhat larger due to not accounted for low-frequency fluctuations that are difficult to treat quantitatively. We consider the symbol size in Fig. 1 as a good estimation of the total error. All measurements have been performed at 25 °C.

References

[Köhler(1993)] W. Köhler, *J. Chem. Phys.*, 1993, **98**, 660.

[Wittko and Köhler(2003)] G. Wittko and W. Köhler, *Philos. Mag.*, 2003, **83**, 1973.

[Köhler and Rossmanith(1995)] W. Köhler and P. Rossmanith, *J. Phys. Chem.*, 1995, **99**, 5838.

Table 1: Measured contrast factors and Soret coefficients

Mixture	x	c	$(\partial n/\partial T)_{p,c}$	$(\partial n/\partial c)_{p,T}$	S_T
			$10^{-4}K^{-1}$	10^{-2}	$10^{-3}K^{-1}$
C_6H_5Br/C_6H_5Cl	0.2001	0.2587±0.0004	-5.38±0.01	3.02±0.09	2.64±0.08
	0.3522	0.4313±0.0004	-5.37±0.01	3.35±0.05	2.49±0.04
	0.4982	0.5807±0.0004	-5.37±0.01	3.64±0.05	2.53±0.04
	0.6471	0.7189±0.0003	-5.36±0.01	3.90±0.08	2.51±0.05
	0.7998	0.8479±0.0003	-5.35±0.01	4.15±0.10	2.74±0.08
C_6H_5Br/C_6H_5F	0.2221	0.3181±0.0004	-5.61±0.01	8.00±0.08	3.78±0.04
	0.4151	0.5369±0.0004	-5.53±0.01	9.52±0.05	3.72±0.02
	0.6004	0.7106±0.0003	-5.48±0.01	10.72±0.08	3.73±0.03
	0.8016	0.8684±0.0003	-5.42±0.01	11.82±0.10	3.70±0.04
C_6H_5Cl/C_6H_5F	0.2014	0.2281±0.0004	-5.63±0.01	5.64±0.10	1.50±0.05
	0.5016	0.5410±0.0004	-5.54±0.01	5.86±0.05	1.48±0.02
	0.7995	0.8237±0.0003	-5.44±0.03	6.04±0.10	1.51±0.04
C_6H_5Br/C_7H_8	0.2506	0.3630±0.0004	-5.59±0.01	5.30±0.03	4.78±0.02
	0.4988	0.6291±0.0004	-5.50±0.01	6.95±0.02	4.42±0.02
	0.7478	0.8348±0.0004	-5.42±0.01	8.22±0.05	4.35±0.03
C_6H_5Cl/C_7H_8	0.2512	0.2907±0.0003	-5.60± 0.01	2.53±0.03	2.59±0.04
	0.5014	0.5513±0.0004	-5.51±0.01	2.79±0.02	2.35±0.02
	0.7413	0.7778±0.0004	-5.46± 0.01	3.02±0.04	2.18±0.04
C_6H_5F/C_7H_8	0.2498	0.2578±0.0003	-5.68±0.01	-2.86±0.02	1.08±0.01
	0.4999	0.5104±0.0004	-5.69±0.02	-3.08±0.01	0.83±0.01
	0.7467	0.7546±0.0004	-5.70±0.01	-3.30±0.02	0.77±0.01
C_6H_5Br/C_6H_{12}	0.2023	0.3212±0.0003	-5.47±0.03	10.19±0.07	2.05±0.02
	0.3035	0.4484±0.0004	-5.44±0.01	12.15±0.05	2.50±0.01
	0.5109	0.6609±0.0004	-5.43±0.02	15.41±0.07	3.18±0.02
	0.6047	0.7405±0.0004	-5.37±0.03	16.63±0.09	3.44±0.03
	0.7939	0.8778±0.0004	-5.35±0.01	18.74±0.1	3.71±0.03
C_6H_5Cl/C_6H_{12}	0.2048	0.2561± 0.0003	-5.50± 0.01	7.73± 0.09	-0.49±0.01
	0.4975	0.5697±0.0004	-5.46±0.01	10.12±0.06	0.46±0.01
	0.6304	0.6953±0.0004	-5.44±0.01	11.08±0.08	0.80±0.01
	0.7982	0.8410±0.0004	-5.42±0.01	12.19±0.13	1.13±0.01
C_6H_5F/C_6H_{12}	0.1986	0.2205±0.0003	-5.58±0.01	2.51±0.10	-2.01± 0.09
	0.3013	0.3299±0.0003	-5.61± 0.01	3.01±0.07	-1.85±0.05
	0.5017	0.5348±0.0004	-5.66± 0.01	3.96±0.04	-1.11±0.02
	0.6050	0.6362±0.0004	-5.67± 0.01	4.43±0.06	-0.61±0.01
	0.7989	0.8194±0.0004	-5.70± 0.01	5.28±0.10	0.024±0.02
	0.8959	0.9076±0.0004	-5.71± 0.01	5.69±0.13	0.48±0.03