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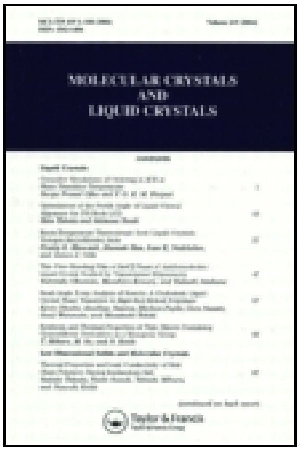

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The Nematic/Isotropic Interfacial Energy: Determination from the Frenkel Relation for Droplet Coalescence

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THE NEMATIC/ISOTROPIC INTERFACIAL ENERGY: DETERMINATION FROM THE FRENKEL RELATION FOR DROPLET COALESCENCE

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According to the theory of Frenkel, the coalescence kinetics of two identical touching droplets is governed by their surface energy, viscosity and size. We show that the Frenkel relation can be used to determine γ_{NI}, the nematic/isotropic interfacial energy, from observations of coalescing nematic droplets in an isotropic matrix. By applying the Frenkel theory to the data of Kahlweit and Ostner for the coalescence of PAP, we obtain $\gamma_{NI} \approx 0.014$ dynes/cm. This value is in much better agreement with other published interfacial energies than the result of the original Rayleigh-Webb analysis of Kahlweit and Ostner, who found an approximate lower limit of $5x10^{-4}$ dyne/cm for γ_{NI}.

The surface energy γ_{NI} at the interface between the nematic and isotropic phases of a liquid crystal is of interest because of its relationship to the orientational order parameter and coherence length in the transition layer between the two phases.[1-4] However, relatively little quantitative information on γ_{NI} has been published.

Murakami,[2] using a statistical mechanical model, computed $\gamma_{NI} \approx 0.01$ to 0.02 dyne/cm for MBBA. Yokayama,

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et al.[3] used the Landau-de Gennes theory to calculate γ_{NI} $\approx$ 0.011 dyne/cm for 5CB. From light scattering measurements, Langevin and Bouchiat[4] determined γ_{NI} = 0.023 for MBBA. Kahlweit and Ostner[5] applied the Rayleigh-Webb theory of oscillating drops to microscopic observations of coalescing nematic droplets to estimate a lower limit of $5x10^{-4}$ dyne/cm for γ_{NI} of PAP (p-azoxyphenetole) at T_{NI}, the nematic-isotropic transition temperature. Recently Smith, et al.[6] applied the Frenkel theory[7] to their observations of coalescing mesophase spherules to obtain a value of $\sim$0.02 dyne/cm for the interfacial energy between the carbonaceous nematic mesophase and isotropic phase of a petroleum pitch. The agreement of this first determination of γ_{NI} for a planar mesophase with values for systems composed of rod-shaped molecules reinforces the view of Langevin and Bouchiat that the nematic-isotropic interface behaves practically the same as the interface between normal liquids having the same densities and viscosities.

The purpose of the present letter is to suggest that the Frenkel theory is better suited for the evaluation of γ_{NI} from coalescence kinetics than the Rayleigh-Webb theory used by Kahlweit and Ostner. We shall test and confirm this view by applying the Frenkel theory to recalculate γ_{NI} for PAP (using the original data of Ref. 5) for comparison with results for other nematogens.

Frenkel derived the following approximate relation for τ, the time constant for coalescence of two identical touching droplets:

$$\tau \approx \frac{\eta R}{\gamma} \quad . \tag{1}$$

Here η is the viscosity, R the droplet radius, and γ the interfacial energy. We obtain values of R and τ for

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coalescing droplets of PAP from Figs. 1 and 2 of Kahlweit and Ostner. Miesowicz[8] has reported viscosity coefficients (η_1, η_2, and η_3) for PAP at 144.4°C, some 23°C below T_{NI}. As Langevin and Bouchiat[9] have shown, the anisotropy of viscosity disappears at T_{NI} so that

$$\eta_1 = \eta_2 = \eta;\ \eta_3 = 0\ . \tag{2}$$

Thus, we may obtain a reasonable estimate of η at T_{NI} by taking the mean of Miesowicz's η_1 and η_2 values. In doing this we are, of course, ignoring the decrease in viscosity due to the 23°C temperature difference. The resultant uncertainty in viscosity should not amount to more than a factor of two, which is acceptable in light of the approximation in the Frenkel relation.[7]

Kahlweit and Ostner report that nematic droplets of PAP with $R \approx 0.013$ cm coalesce almost completely in 0.045 s. From the Miesowicz viscosities we obtain $\eta \approx (\eta_1 + \eta_2)/2 = (0.013 + 0.083)/2 = 0.048$ poise.* Substituting these values into Eq. 1 gives

$$\gamma_{NI} \approx 0.014 \text{ dyne/cm.}$$

The good agreement of this result with the above theoretical and experimental values for nematic/isotropic interfacial energies of other systems (composed of rod-shaped and planar molecules) confirms the validity of the Frenkel relation and strongly supports its usefuless in determining γ_{NI} for other nematogens using optical microscopy, a relatively simple and straightforward procedure.

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*de Jeu[10] has pointed out a difficulty with the notation of Miesowicz as well as a small error in one of his values. However, these discrepancies do not affect the present results to an appreciable degree.

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