Brilliant total reflection from a liquid-liquid interface

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Contributed by Joel H. Hildebrand, September 13, 1978

ABSTRACT In this paper I describe total reflection of great brilliancy that seems to indicate that an interface can be very thin.

In 1969, Huang and Webb (1) published an exhaustive study of a diffuse interface in a critical fluid mixture of cyclohexane and methanol.

In the study described here, the two liquids were carbon tetrachloride and perfluoroethylcyclohexane ($\text{c-C}_6\text{F}_{11}\text{C}_2\text{F}_5$) which Lamoreaux and I (2) used for demonstrating the enormous increase in the range of thermal fluctuations as the critical temperature is approached from above (see also ref. 3). The density of CCl_4 at 25°C is 1.594 g-cm^{-3} and its refractive index at the D line is 1.461 (4). The density of $\text{c-C}_6\text{F}_{11}\text{C}_2\text{F}_5$ at 25°C is 1.771 g-cm^{-3} and its index of refraction is 1.277 (5). The critical temperature is 24.70°C . These liquid phases present a rare case of an upper layer that has a much larger refractive index than the lower one, and therefore light can be totally reflected at the upper side of the interface at a considerable angle from the horizontal. Of course, each component is somewhat soluble in the other. Hence, the two refractive indices approach one another as the temperature approaches the critical temperature; but the solubility loop is nearly flat at the top, and volume fractions and mole fractions differ by nearly 15% at 2°C below the critical temperature.

In order to see a linear reflected beam, I asked our skillful glassblower to make for me a glass bottle with plane sides and a finely ground stopper. It was 12 cm tall with rectangular cross section $2.5 \times 5\text{ cm}$.

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The light from a 40-W frosted bulb mounted 20 cm from the bottle and a little above the level of the interface was reflected at an angle of about 15° above horizontal when the temperature of the interface was 23°C . The reflected beam was not visibly less bright than light reflected from a silvered mirror held horizontal beside the bottle at the level of the interface. That an interface that is nearly invisible from other angles can show a total reflection from a 40-W bulb close to it that is virtually as bright as the reflection from a silver mirror amazes everyone who has seen it.

As temperature slowly approached the critical temperature, the beam was reflected at a decreasing angle and thermal fluctuations became long enough to scatter blue wavelengths; the beam became tinged with red before it disappeared.

Those who have investigated the structure of critical interfaces have, so far as I have seen, used at least one polar component; my two liquids have nonpolar, fairly symmetrical molecules. It seems to me that the interface must be regarded as exceedingly thin. I hope to hear opinions on this point.

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5. Lovelace, A. M., Rausch, D. A. & Postelnek, W. (1958) *Aliphatic Fluorine Compounds*, American Chemical Society Monograph Series (Reinhold, New York).

Abbreviation: $\text{c-C}_6\text{F}_{11}\text{C}_2\text{F}_5$, perfluoroethylcyclohexane.