

or does not apply will be published later.

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CRITICAL OPALESCENCE OF LIQUID SODIUM-LITHIUM MIXTURES

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The critical opalescence of binary liquid mixtures near the critical demixing temperature T_c has been investigated by means of light or x-ray scattering for various systems (see Brumberger¹ for a review of the experimental literature). The observed increase in scattered intensity as T approaches T_c from the one-phase region is due to the increasing ease with which concentration fluctuations (and consequently fluctuations in electron density or refractive index) occur, and to the rapidly increasing long-range correlation between these fluctuations. Various theoretical treatments have been given.²⁻⁵ Our purpose in this Letter is to report the experimental detection of what we believe to be critical opalescence in a type of system where this phenomenon has not previously been observed, namely a binary liquid-metal system, Na-Li.

The small-angle x-ray scattering of the sodium-lithium system in the critical region, at a composition of 58 with respect to Na, was investigated using a Kratky camera,⁶ MoK α radiation monochromatized by filter and pulse-height discrimination, and a monitor detector to eliminate intensity variations due to any source instabilities. The liquid metals were confined in a high-temperature sample cell described in detail elsewhere.⁷ The cell was filled directly from a mixing crucible where samples were prepared, melted, homogenized, and freed of any undissolved impurities *in situ* under high-purity argon. Sample thickness in the x-ray path was 3.62 mm. Widely different values of T_c have been reported^{8,9}; for this reason, a series of angular scans were made over a substantial temperature range, 430 to 280°C.

The temperature of the sample was initially allowed to decrease at the rate of 1°C per 30 min; when the desired temperature was reached, the sample was allowed to equilibrate for 30 min, during which interval the intensity at a fixed angle was read every 2 min. In the subsequent interval, a complete angular intensity scan was made, the sample temperature was then lowered to the next desired value, and the procedure was repeated. Temperatures were measured by means of a Wenner potentiometer and iron-iron/Constantan thermocouples calibrated against National Bureau of Standards certified, mercury-in-glass thermometers of the appropriate range. The temperature of the sample could be controlled to $\pm 0.05^\circ\text{C}$ or better, and was uniform over the irradiated region to this precision.

Figure 1 shows a typical plot of scattered intensity (counts min⁻¹) at a fixed scattering angle θ of 10^{-3} rad (an equivalent Bragg spacing of about 700 Å) versus temperature. A substantial increase in intensity in a range of about 10°C above 301°C and a sudden sharp drop below this temperature are observed. From this graph one would estimate that T_c is $\sim 301^\circ\text{C}$; this is somewhat below the value of 303°C shown for this composition by Kanda, Faxon, and Keller,⁹ but in good agreement in view of the experimental uncertainties and slight differences in sample purity, composition, and technique.

The general features of the effect are reproducible; some variations in T_c , intensity distribution, and temperature range of the effect are observed from sample to sample. These are primarily due to slight variations in purity, sample thickness, and composition. The

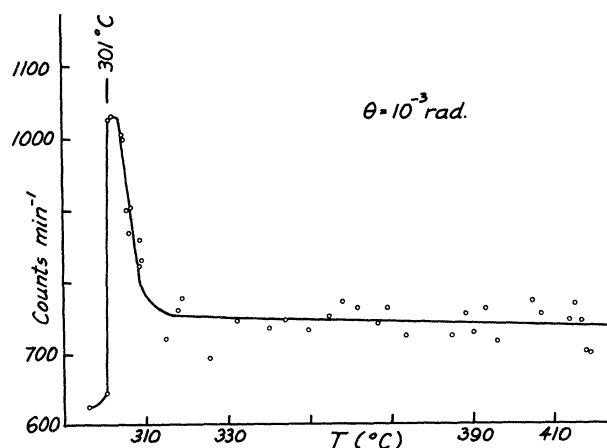


FIG. 1. Plot of intensity (uncorrected for slit smearing or background) versus temperature for a scattering angle of 0.001 rad. Background is nearly constant with T above T_C and therefore does not affect the shape of this curve.

temperature range over which the intensity increases compares roughly with the range reported by Münster and Sagel for the opalescence of the solid solution Al-Zn.¹⁰

Calculations using the phase diagram and density data of Ref. 9 and the known geometry of the sample cell have convinced us that it is not possible to account for the observations above 301°C on the basis of changes in x-ray absorption occurring in the neighborhood of

the phase-separation temperature because of the formation of two liquid layers whose volumes vary with T .

Detailed data and an analysis of our results on the basis of existing theories will be reported in a future, more extensive publication.

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OPTICAL THIRD-HARMONIC GENERATION IN GASES

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Experimental optical third-harmonic coefficients for inert gases are presented and compared with experimental Kerr coefficients and theoretical dc hyperpolarizabilities.

We wish to report the observation of optical third-harmonic generation (THG) in gases, particularly helium, neon, argon, krypton, and xenon. THG in crystals, glasses, and liquids has been reported previously by Maker, Terhune, and Savage.¹ The motive for studying these gases is that the nonlinear susceptibilities are relatively amenable to quantum-mechanical calculation. Interesting features of the propagation of third-harmonic radiation have also become apparent.

A schematic diagram of the apparatus is shown

in Fig. 1. A Q-switched ruby laser (Lear-Siegler LS 100) provided a fundamental light pulse (6943 Å) of 50-nsec duration and about 2-MW peak power. The signal from a photodiode monitoring the fundamental light was displayed on one beam of a dual-beam oscilloscope (Tektronix Type 551). The laser beam passed through a red filter to attenuate stray flash-tube light and was focused in front of the glass entrance window of the gas cell. All succeeding optical components were made of quartz to pass the ultraviolet third-harmonic radiation (2314 Å).