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# In-Plane Structure of the Liquid-Vapor Interface of an Alloy: A Grazing Incidence X-ray Diffraction Study of Bismuth:Gallium

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**The liquid-vapor interface of a bismuth-gallium mixture (0.2 percent bismuth and 99.8 percent gallium) at 36°C has been studied by grazing incidence x-ray diffraction. The data show, in agreement with thermodynamic arguments, that bismuth is heavily concentrated in the liquid-vapor interface. The x-ray diffraction data are interpreted with the assistance of a simple model that represents the interface as a partial monolayer of bismuth. This analysis leads to the conclusion that the bismuth concentration in the interface is about 80 percent, that there is no significant mixing of gallium and bismuth in the interface, and that the structure function of the interfacial bismuth is like that of supercooled bulk liquid bismuth.**

The liquid-vapor interface is a region that separates two fluids that have very different densities. As such, it is an interesting vehicle for the study of inhomogeneous fluids. Consider first a pure liquid. A principal goal of the study of inhomogeneous fluids is the determination of the relations between the anisotropy associated with the density gradient, intermolecular interactions, the longitudinal structure of the liquid (parallel to the density gradient), and the transverse structure of the liquid (perpendicular to the density gradient). A liquid mixture differs from a pure liquid in that the equilibrium concentrations of the components in the homogeneous bulk and in the liquid-vapor interface will, typically, be different; we expect this segregation of components to alter the structure of that interface.

Most investigations of the structure of the liquid-vapor interface have been concerned with the longitudinal density distribution. In a simple dielectric liquid, such as Ar (or water), both theory and experiment agree that along the normal to the liquid-vapor interface the density decreases smoothly, without subsidiary structure, from the liquid density to the vapor density (*1*). On the other hand, for a liquid metal, theory predicts (*2, 3*) and experiment very weakly confirms (*4, 5*) that the longitudinal density distribution in the liquid-vapor interface has oscillations with a period of an atomic diameter, extending about three atomic diameters into the bulk. For a simple dielectric liquid mixture, theory predicts that the longitudinal density distribution of the component that is in excess in the liquid-vapor interface falls smoothly with distance into the bulk, the range of the decay being several atomic diameters. In contrast, in a liquid metal mixture, such as Cs:Na, theory predicts that the component that concentrates in the liquid-vapor interface forms a nearly pure monolayer, that the immediately adjacent liquid on the bulk side of the interface is depleted in the surface-active component for about an atomic diameter, and that (very nearly) the bulk concentration is reached at a distance of about two atomic diameters into the bulk (*3*).

Although it has been studied even less than the longitudinal density distribution, the transverse structure of the liquid-vapor interface has been the subject of a number of theoretical and experimental studies. Consider first liquid metals. It has been shown, both experimentally (*6, 7*) and theoretically (*8*), that the transverse structure function of the liquid-vapor interface of a pure metal is very similar to the bulk structure function despite the oscillatory nature of the longitudinal density distribution in the liquid-vapor interface. The near identity of the transverse structure function in the liquid metal–vapor interface and the bulk liquid metal structure function characterize a situation very different from that found in crystalline media. In the latter case the surface of the crystal is usually reconstructed and has a different atomic arrangement than does the bulk crystal. On the other hand, when applied to a simple dielectric liquid, the theoretical approach used to describe the liquid metal–vapor interface leads to the prediction that the transverse structure function in the liquid-vapor interface will be different from the bulk liquid structure function. There are, as yet, no experimental tests of this prediction.

This report describes the results of a study of the transverse structure function of the liquid-vapor interface of a Bi:Ga alloy with a bulk composition of 0.2% Bi and 99.8% Ga. In this system the Bi which

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segregates in the liquid-vapor interface is present at a concentration that greatly exceeds the bulk solubility at the selected temperature. Indeed, at the temperature of the system pure Bi would be solid. What, then, is the structure of the Bi that segregates in the liquid-vapor interface of this system?

If the liquid alloy is an ideal binary mixture, and the surface tensions ($\gamma_i$), bulk mole fractions ($x_i$), and molar areas ($A_i$) of the components are known, the surface mole fractions ($y_i$) can be calculated from the Gibbs adsorption isotherm:

$$\gamma_M = \gamma_1 + \frac{RT}{A_1}\ln(y_1/x_1) = \gamma_2 + \frac{RT}{A_2}\ln(y_2/x_2) \qquad (1)$$

The application of Eq. 1 to the 0.2% Bi:99.8% Ga alloy leads to the prediction that the mole fraction of Bi in the liquid-vapor interface is 0.76. We shall see that the predicted surface enhancement of Bi is, within experimental error, the same as that deduced from our analysis of the grazing incidence x-ray diffraction data.

We used the same sample chamber and diffractometer as in our study of the transverse structure function of the liquid-vapor interface of Ga (7). The calibration of the scattering angles was determined by diffraction from Si powder obtained from the National Institute of Standards and Technology. We prepared the alloy sample by heating ~500 g of Ga (99.9999% pure) to about 60°C and stirring in 3.3 g of Bi powder (99.99% pure). We prepared the chamber to receive the sample by pumping, with baking at 110°C, down to a total pressure of $6 \times 10^{-10}$ torr with less than $2 \times 10^{-13}$ torr $O_2$ and $1 \times 10^{-12}$ torr $H_2O$ vapor. The sample was introduced into the chamber at a temperature of about 100°C because at lower temperatures it did not flow freely, perhaps as a result of freezing inside the capillary. Immediately after introduction of the sample the water vapor pressure rose to about $8 \times 10^{-8}$ torr. Although the pressure dropped to $9.9 \times 10^{-9}$ torr after 2 hours, the system with sample in situ was baked for a day to remove the residual adsorbed water vapor. After the residual gas analyzer attached to the sample chamber showed a partial pressure of $O_2$ less than $2 \times 10^{-13}$ torr and $H_2O$ vapor less than $1 \times 10^{-12}$ torr, the temperature of the sample was lowered to 34°C and the surface swept clean with a stainless steel arm. [In our study of the liquid-vapor interface of Ga (7) we studied an unswept surface, then a swept surface using the same equipment. There was no difference between the two surfaces that we could detect. We take this as evidence that our equipment deposits clean metal into the chamber and that no detectable oxidation of the surface occurs within a week under a pressure of $2 \times 10^{-13}$ torr of $O_2$ (with a total pressure of $1 \times 10^{-9}$ torr).] A wrinkled metallic film was seen to form as the surface layer was removed, showing none of the whiteness that characterizes oxidized surfaces; warming the sample to 36°C rendered the liquid-vapor interface smooth, shiny, and fluid.

Figures 1 and 2 display raw transverse scattering data and bulk liquid scattering data, respectively. It is relevant to make two comments concerning these data. First, the arrangements of the incident beam and the detector are nearly identical for the two experiments because the change in angle of incidence is only from 4 mrad to 8 mrad. The x-ray penetration depth at 4 mrad is about 30 Å, whereas at 8 mrad, which is above the critical angle for reflection, it is much greater and is determined by the absorption length in liquid Ga. The efficiencies of signal detection are the same for the two experiments. Second, the x-rays are strongly polarized parallel to the plane of the liquid-vapor interface, which greatly reduces the scattered x-ray intensity between $k = 5$ Å$^{-1}$ and $k = 7$ Å$^{-1}$. The raw data displayed show that the structure of the liquid-vapor interface of the alloy is profoundly different from that of bulk. The presence of the peak near $k = 2$ Å$^{-1}$ in the transverse scattering data suggests the presence of liquid Bi at the surface.

A complete characterization of the structure of a binary mixture requires three structure functions. For an alloy A:B the conventional choices for these functions are the three partial structure factors that describe the A-A, B-B, and A-B correlations in the liquid. Then the total structure factor, $S(Q)$, is defined by

$$S(Q) = W_{AA}S_{AA}(Q) + W_{BB}S_{BB}(Q) + 2W_{AB}S_{AB}(Q) \qquad (2)$$

where

$$W_{\alpha\beta} = \frac{x_\alpha x_\beta f_\alpha f_\beta}{\langle x_\alpha f_\alpha + x_\beta f_\beta \rangle} \qquad (3)$$

where $f_j$ is the atomic structure function of species $i$ and $x_i$ is the mole fraction of species $i$. Clearly, the data available are insufficient for a unique determination of the three partial structure functions. However, the Bi concentration in the alloy is so low that the bulk structure function can be determined from the bulk scattering data if the alloy is treated as a pure metal. The results displayed in Fig. 3 show that the structure function of the very dilute bulk liquid alloy is, indeed, sensibly the same as that of pure liquid Ga.

Our analysis of the transverse scattering

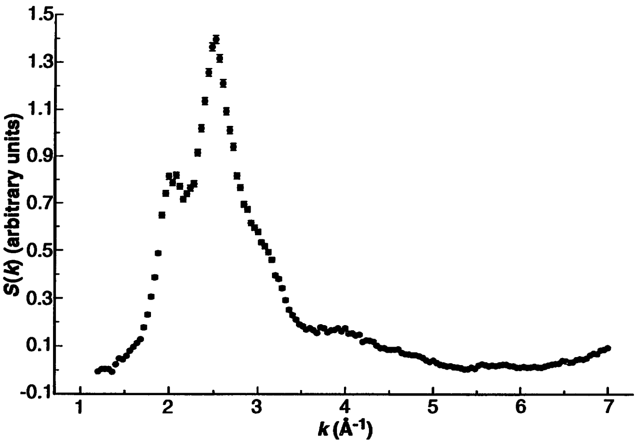

**Fig. 1.** The surface structure function data, incident angle of 4 mrad, shown without any corrections. In this figure, and in all subsequent figures, all of the data points shown include error bars. Where these error bars are not visible they are the same size or smaller than the symbol used to represent the the data point.

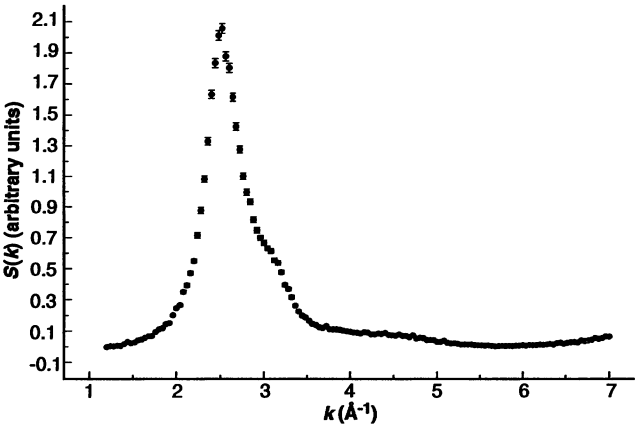

**Fig. 2.** The bulk structure function data, incident angle of 8 mrad, shown without any corrections.

data is based on the assumption that the excess Bi in the liquid-vapor interface forms a partial monolayer. We assume, in analogy with the result of the Harris, Gryko, and Rice calculation (*3*) of the structure of the liquid-vapor interface of Cs:Na, that immediately beneath the Bi monolayer the alloy has the bulk composition. Because the x-ray beam incident on the sample generates an evanescent wave in the alloy, the observed transverse scattering signal has contributions from both the liquid-vapor interface and the bulk alloy. The form and amplitude of the decaying x-ray field in the bulk liquid are known, as is the bulk structure function, so the latter contribution can easily be accounted for in the reduction of the experimental data. A key element in our analysis is the theoretical prediction (*8*) and experimental verification (*6*, *7*) that the transverse structure function in the liquid metal–vapor interface is sensibly identical with the structure function of the bulk liquid.

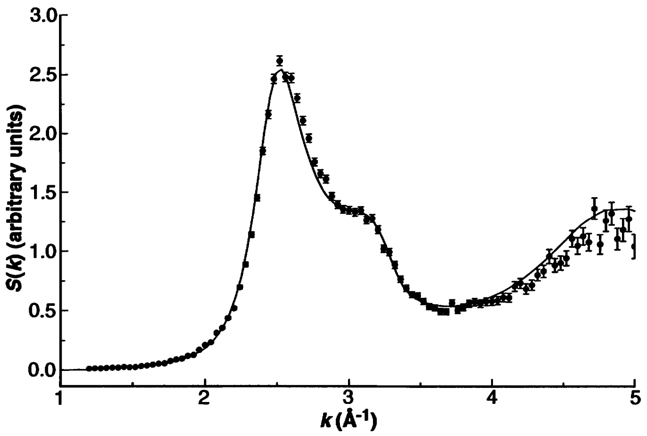

**Fig. 3.** The bulk structure function data corrected for polarization, diffractometer geometry, atomic scattering function, and inelastic scattering (points), compared to the Ga structure function determined by Narten (*10*) (solid line).

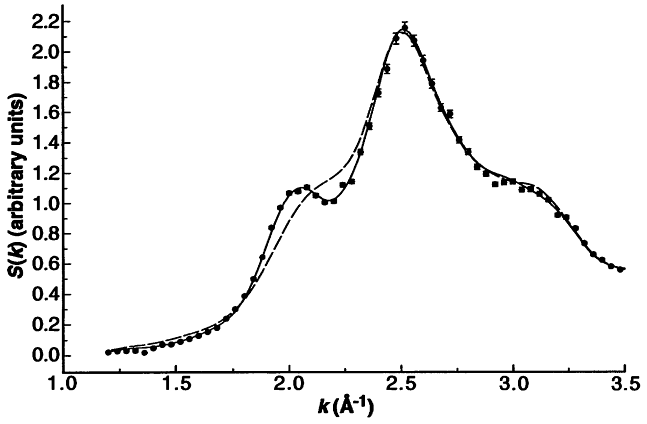

**Fig. 4.** The two fits of our data to a linear combination of Ga and Bi structure functions. The points are our data adjusted for all relevant factors, the dashed line is a combination of Narten's Ga structure function (*10*) and Dahlborg and Davidovic's (293°C) Bi structure function (*9*), and the solid line is Narten's Ga structure function combined with a modified Bi structure function (as described in the text). Note the qualitative failure of the fit with the 293°C bulk liquid structure function.

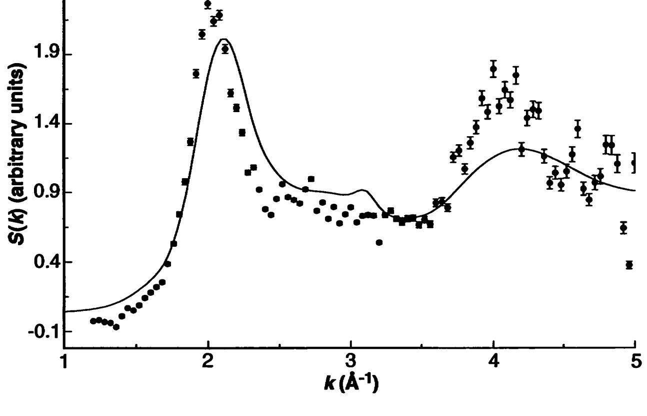

**Fig. 5.** The structure function remaining after the Ga contribution estimated by the solid line fit in Fig. 4 is subtracted from the data; (points) our surface Bi *S*(*k*) at 36°C; (solid line) bulk Bi *S*(*k*) at 293°C. The data show peaks that are taller and narrower and are shifted to lower *k* compared to the 293°C bulk liquid structure function.

Adopting the model described, we proceeded as follows. First, the raw scattering data were transformed by accounting for viewing geometry, polarization, and so forth, none of which are dependent on the composition of the alloy. Then we fit the transformed data with a linear combination of partial structure factors, using the experimental data of Dahlborg and Davidovic (*9*) for $S_{\mathrm{BiBi}}(Q)$, the data of Narten (*10*) for $S_{\mathrm{GaGa}}(Q)$, and a synthetic $S_{\mathrm{BiGa}}(Q)$ constructed by averaging the normalized and reduced structure functions for pure Bi and pure Ga; the reduced structure functions were defined with respect to the momentum transfer characteristic of the first peak of each $S(Q)$. The least squares fit of the linear combination shown in Eq. 2 to the data (Fig. 4, dashed line) with the partial structure functions defined as stated is a combination of Bi-Bi and Ga-Ga structure functions, with no contribution from the Bi-Ga structure function. But, is the bulk Bi-Bi structure function appropriate for the description of putatively liquid Bi at a temperature which is 271°C below its normal freezing point? Perhaps a more appropriate reference liquid structure function is that for supercooled Bi. Indeed, an improved fit to the observed transverse structure function can be generated (Fig. 4, solid line) if we modify the Bi transverse structure function in the interface by shifting the first peak of $S_{\mathrm{BiBi}}(Q)$ by 4%, to a reduced momentum transfer of 0.96, and then narrowing and amplifying the first peak while preserving its area. A good fit was obtained in the region of the first peak when the peak width was decreased by 20% and the peak height increased by 20%, consistent with the changes in $S(Q)$ induced by supercooling a liquid. After estimating the relative Ga and Bi contributions in the region from $k = 1$ to 3.5 Å$^{-1}$ with the modified structure factor, we subtract the estimated Ga contribution from our data, thereby generating a residual structure function. In Fig. 5 we compare that residual structure function with the structure function of liquid Bi near the melting point. We take the residual surface structure function as descriptive of the atomic correlations in a segregated Bi monolayer in the liquid-vapor interface of the alloy.

To relate the weightings of the partial structure functions in the best fit to the observed transverse structure function to the concentrations of species in the interface, we carried out a three-part analysis. First, we normalized the areas of all the partial structure functions, which permits calculation of the relative contributions to the observed scattering from Bi and Ga, including the scattering which arises from the penetration of the evanescent wave into the bulk liquid. Second, we assumed that the partial monolayer of Bi in the liquid-vapor interface is supported on pure Ga. Third, from the known amplitude and penetration of the evanescent electric field in the bulk liquid we calculated the relative illumination by the x-rays of the two species in our model. To carry out these calculations we must know the densities of the several species. Nothing is known of the density of the Bi monolayer, but the form of the transverse structure function suggests that it might be in a supercooled liquid state, so we took the density to be 10.0 g/ml, which is between the solid- and normal liquid-state densities. The results of these calculations are $y_{\mathrm{Bi}} = 0.80$ and $y_{\mathrm{Ga}} = 0.20$. The surface concentration of the segregated Bi determined in this fashion is the same as that predicted from the Gibbs adsorption isotherm and the assumption that the binary alloy is an ideal mixture.

The raw data obtained from our grazing incidence x-ray diffraction study of the liquid-vapor interface of the 0.2%Bi:99.8%Ga alloy clearly show that the transverse structure function of the interface is different from the structure function of the bulk liquid. The qualitative character of the difference observed is consistent with a large excess concentration of Bi in the liquid-vapor interface. A simple but plausible model leads to the conclusion that the mole fraction of Bi in the liquid-vapor interface of the alloy is 0.80, in good agreement with the value predicted from the Gibbs adsorption isotherm. We do not have a good way of testing the validity of the quantitative details of the results of our analysis, but we are confident that the qualitative aspects of our observations are correct. These are (i) that Bi is strongly concentrated at the liquid-vapor interface of dilute liquid Bi:Ga and (ii) that the segregated Bi in the interface is in a liquid state. To unambiguously determine the transverse structure function of the excess Bi in the liquid-vapor interface it is necessary to carry out three experiments under conditions such that the scattering powers of the Bi and Ga atoms are different in each experiment. In principle, these conditions could be generated by taking advantage of the anomalous dispersion of x-rays near an absorption edge. We believe the necessary experiments should be attempted.

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# Pattern Recognition in Coupled Chemical Kinetic Systems

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**A network of open, bistable reaction systems coupled by mass transfer is simulated. The mass transfer rates are determined by a Hebb-type rule, and the programmable network can store patterns of high and low concentrations in each bistable system. In a parallel computation, the network recognizes patterns similar, but not necessarily identical, to stored patterns.**

We have offered suggestions for the chemical implementation of computing machines (*1–3*): the chemical coupling of reaction mechanisms (*4*) far from equilibrium whose stationary states have properties of a McCullouch-Pitts neuron (*5*) led to the construction of logic gates, a finite state machine that generalizes to a universal Turing machine (*6*), and a parallel neural network computer. In this report, we describe a class of parallel chemical computers based on chemical kinetics systems with multiple stable stationary states coupled by mass transfer. We demonstrate the computational capacity of such a network by storing patterns in it and solving pattern recognition problems.

Consider a set of chemical reactions that occur homogeneously in an enclosure, which may be a cell compartment, a neuron, or a stirred reaction vessel. Each enclosure is an open system: reactants enter, and products, intermediates, and unreacted material exit. We use a set of chemical reactions that is bistable. There are many reaction mechanisms and physical processes in which bistability is known, including enzymatic reactions (*7*) and neuronal response (*8, 9*). For simplicity of illustration, we use an inorganic chemical reaction, the iodate–arsenous acid reaction (*10, 11*)

$$\mathrm{IO_3^-} + 3\mathrm{H_3AsO_3} \rightleftharpoons \mathrm{I^-} + 3\mathrm{H_3AsO_4} \qquad (1)$$

for which the kinetics are adequately described by the temporal variation of a single variable, the concentration of $\mathrm{I^-}$ ($[\mathrm{I^-}]$). Bistability occurs in this isothermal reaction: for a given range of input flows of reactants (given constraints), the homogeneous reaction system at steady state may have either a high or low $[\mathrm{I^-}]$.

The systems communicate with each other by mass transfer; either diffusional, in which case it is reciprocal, or active transport, in which case we require it to be reciprocal. Although all the different chemical species are transported, the temporal evolution of the *i*th system ($i = 1, \ldots, N$) in the network is adequately described by its $[\mathrm{I^-}]$ (*11–13*)

$$\frac{d[\mathrm{I^-}]_i}{dt} = -k_\mathrm{B}[\mathrm{I^-}]_i^3 + \{k_\mathrm{B}([\mathrm{I^-}]_0 + [\mathrm{IO_3^-}]_0) - k_\mathrm{A}\}[\mathrm{I^-}]_i^2 + \{k_\mathrm{A}([\mathrm{I^-}]_0 + [\mathrm{IO_3^-}]_0) - k\}[\mathrm{I^-}]_i + k[\mathrm{I^-}]_0 + \lambda \sum_{j \neq i} \mathrm{k_{ij}}([\mathrm{I^-}]_j - [\mathrm{I^-}]_i) \qquad (2)$$

in which $[\mathrm{I^-}]_0$ and $[\mathrm{IO_3^-}]_0$ are, respectively, the concentrations of iodide and iodate in the reactant flows that maintain each system away from equilibrium, $k_\mathrm{A}$ and $k_\mathrm{B}$ are effective rate coefficients, and $k$ is the reactant flow rate. The mass transfer coefficients, $k_{ji} = k_{ij} \geq 0$, are reciprocal and are chosen by a Hebbian rule (*14*), and $\lambda \geq 0$ is a scaling parameter that is chosen such that the final state of each system is influenced by the other systems but not so strongly that the network is always driven to homogeneity. There exists a Liapunov function for chemical systems described by Eq. 2 (*13*), which is an evolution criterion toward stable stationary states (analogous to the Gibbs free energy of a system approaching equilibrium). If each system has only one stable stationary state, then the only stable stationary state of *N* coupled systems

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