Wetting, spreading and Marangoni convection in gallium–bismuth alloys

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Abstract.   The present study reports several novel results pertinent to the wetting transition in Ga–Bi. These results show that complete wetting depends crucially on the boundary conditions between the liquid sample and the container. If liquid Ga does not wet the container wall a thick Bi film of about 50 μ spreads over the Ga surface at nearly room temperature due to the Marangoni effect. If Ga wets the wall the wetting transition occurs near to the monotectic temperature of this system. The interfacial tension inferred from the surface light scattering data displays non-monotonic behaviour on heating.

1. Introduction

In common practice a liquid drop is supposed to wet a solid substrate when it spreads over the substrate. However in a system comprising two mutually soluble components wetting would mean the covering of one component in a liquid state by a film of the second component if the surface energy of this complex interface is reduced. In this case wetting reflects an interplay between irreversible surface and reversible bulk thermodynamics.

Several recent experimental and theoretical papers deal with the wetting behaviour in liquid metal alloys [1–3] (and references therein). The authors use different experimental methods to probe the interface of two component alloys of gallium–bismuth and gallium–lead. Despite the difference in instrumental approaches, nearly all authors postulate the appearance of a thin layer (50 to 200 Å) of the surface active component, i.e. Bi or Pb on top of liquid Ga that happens at some critical temperature given by the bulk phase diagram. Only one group use microscopic samples of a typical size about 10 μ [3] while the others explore the samples of some cm size [1, 2]. The authors of [3] renew their previous results reporting adsorption transition in the form of a Pb monolayer in the supercooled (about 50 K lower than the Ga crystallization point) and very dilute Ga–Pb alloys. They refer to this effect as the 'prewetting line' although the Gibbs adsorption equation was explicitly used in their calculations of the surface concentration of Pb on the supercooled liquid Ga. An interesting feature (in the author's opinion) is the experimental finding of a clearly decreasing surface Pb concentration on heating in dilute Ga–Pb alloys. This dependence was observed for different bulk compositions on both the supercooled samples and samples heated above the Ga melting point. Put another way this finding implies a *dewetting* above the prewetting temperature in Ga–Pb (using the original terminology of [3]). The same phenomenon though at a higher temperature has been demonstrated on Ga–Bi in the papers of the Chicago group [4]. We shall return to this point in section 4 where the same tendency will be inferred from our light scattering data.

We will show that a new effect of Marangoni convection on the wetting transition is essential for the surfaces of cm size. Furthermore, a Bi film on Ga is found in the present experiment to be rather macroscopic, clearly detectable by eye and the film thickness measured on solidified samples is about 50 μ. This result strikingly differs from all previously reported in the bulk of the literature. In addition we will demonstrate that the temperature of wetting transition strongly depends on the boundary conditions between a liquid sample and a container. Thus for non-wetting between liquid Ga and a container wall the 'critical temperature' is slightly above the Ga melting point. By contrast the wetting between the sample and the wall shifts the appearance of a thick Bi film to some temperature interval very close to the liquid–liquid coexistence.

2. Experiment

It is essential to have a clean oxide free surface of both components: gallium and bismuth. The Bi surface was prepared first. A batch of about 45 g (99.99, Alfa) was melted and dropped under a high vacuum from a glass syringe to a polished Ti trough of diameter 50 mm. The complete Bi layer was less than 3 mm thick. After solidification its surface was perfectly shiny with typical crystalline planes ensuring no macroscopic oxide film on it. Then liquid Ga (about 65 g 99.999, Alfa) was dropped using the same technique. In principle, a Ga drop wets a Bi-substrate only partially: the contact angle at the Ga–Bi interface is smaller than the angle between Ga and the Ti substrate (as judged by eye). However Ga does not spread along the Bi surface even on heating up to 100 °C. Therefore the complete Ga layer is strongly convex near the wall and is about 6 mm thick in the middle, see figure 1. The sample was sputtered with an Ar$^+$ beam having the energy 2 keV at 10^{-5} Torr integral pressure of argon. Then it was heated in a vacuum of about 3×10^{-8} Torr at a constant heating rate 0.25 °C min^{-1}.
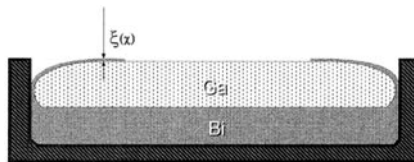
Figure 1. Experimental geometry showing the spreading of a Bi film along the Ga surface. The film whose thickness $\xi(x)$ varies along the interface can flow only if there is no wetting between Ga and container.

We tried to collect the capillary wave spectrum at a fixed wavenumber q for different temperatures. Our light scattering technique is described in detail elsewhere [5–7]. In brief, a beam from a 7 mW He–Ne laser (TEM$_{00}$, $\lambda = 632$ nm) fell on the liquid surface. Small-angle scattered light was optically mixed (on a photodetector) with a portion of the original beam, providing all the necessary conditions for optical heterodyning. The output of a photomultiplier was modulated at the propagation frequency of a capillary wave with the selected wavenumber q. The spectral representation of the signal was recorded in the frequency domain with a spectrum analyser. The whole apparatus was placed on an optical table, vibration isolation being provided by four pressurized air cylinders in the legs.

Just above the starting point ($T \sim 40$ °C) we have seen a strong convective motion on the Ga surface. This convection appeared in the form of non-stationary vortices that moved very fast in the surface plane. The motion was detected by means of light scattered by oxide particles caught by the surface flow. Unfortunately no technique was available to measure the surface velocity which appeared to be more than 1 cm s^{-1}. This is a clear signature of Marangoni convection stemming from the surface tension gradient along the curved Ga surface. The main reason for this gradient is the difference between the tension at the upper Ga surface and

at the lower one ($\delta\gamma_{Ga/Bi}$) which arises from the concentration boundary layer at the Ga–Bi interface. Concentration of Bi in this layer changes rapidly from ∞ to some value given by the thermodynamic phase diagram at a fixed temperature. Put another way, there is a thin Bi-rich film whose thickness varies along the interface near the wall. It is clear that such a film can be unstable (with respect to disturbances of temperature or concentration) and finally can spread over the Ga surface which is schematically drawn in figure 1. Moreover, $\nabla\gamma_{Ga/Bi}$ along the interface sustains Marangoni flow, which in turn is responsible for the transport of Bi from the region having a thicker film to regions covered by a Bi monolayer. It should be stressed that Marangoni convection is driven by the concentration gradient of the tension but not by the temperature gradient. This point is supported by observations on pure gallium placed in the same chamber and heated to the same temperature. After sputtering the liquid surface the island of oxide particles was motionless and stuck to the wall at a fixed position.

In the presence of strong motion together with mechanical vibrations propagating in a thick Ga layer the collection of the light scattering data is nearly impossible. Even if capillary wave spectra were collected the evaluation of the surface tension from the roots of the dispersion equation would be incorrect. The point is that the surface motion changes the dispersion equation written originally for waves of small amplitude. We present no measured data in this case.

On heating Bi dissolves in liquid Ga and its bulk concentration at each temperature is determined by a phase diagram. At the same time we expect the thickness of the Bi film to grow upon heating due to a decrease in the surface energy of the Ga–Bi interface. The film thickness cannot be measured *in situ* in the present study. However the presence of a macroscopic Bi film was detected using visual observations on cooling the sample. Bi film experiences a phase transition (liquid to solid) at some temperature distinct from the crystallization point of liquid Ga. Hence in some T-interval a solid Bi film floats on the liquid substrate. Such a film is easily detectable by eye or by means of strongly enhanced scattering. The temperature of phase transition strongly depends on film thickness: the thicker the film the higher is its crystallization point. To illustrate this we mention that a sample heated to $150\,^\circ$C displayed a solid film on cooling to about $80\,^\circ$C, whereas a solid film appeared at $T \sim 40\,^\circ$C at the surface of the other sample (of the same composition) initially heated to only $60\,^\circ$C. In both experiments a solid Bi film floating on the liquid Ga substrate was detected at T above the crystallization point of Ga.

These observations could be complemented by an experiment utilizing a somewhat different geometry. Instead of a continuous Bi film on the bottom two Bi droplets (each of about 1 g) were placed approximately along a container diameter. Then a Ga layer (from the batch of 60 g) was prepared as described above. Note that the present sample composition was chosen to be far away from that composition ($\sim$20 wt% Bi) which is required for the monotectic point $T_m = 222\,^\circ$C. This composition is necessary for achievement of critical wetting as argued in [1, 2]. The sample was heated under vacuum at T of about $50\,^\circ$C. After sputtering the surface a laminar flow was observed nearly along the line between the Bi droplets. After keeping the sample for one night (at $T \sim 50\,^\circ$C) and subsequent solidification the surface was completely covered with a macroscopic Bi film of a typical yellow colour. All these observations confirm that neither the monotectic temperature nor the concentration at this point defined by the bulk phase diagram is important for the surface wetting. In contrast this phenomenon crucially depends on the experimental geometry or a characteristic time which is a typical signature of a convective transfer of Bi along the surface (see below).

In all our experiments the Bi film was macroscopically thick having a colour clearly different from the colour of solid Ga. The thickness ($50\,\mu$) was measured with raster electron microscopy applied to a cross section (mm-sized) cut from a solid sample. We do not pretend

that our result is an exact replica of the thickness of liquid film: it may change on cooling the sample due to exchange of Bi between the surface and liquid bulk. However it is difficult to see how it could change by the several orders of magnitude by which our value differs from those reported in the bulk of the literature.

3. Spreading of Bi film at liquid Ga surface

In the following text we will explain the physical origin of the anomalously thick Bi film on liquid Ga. The surface tension at the boundary between liquid gallium substrate and vacuum is modified by a bismuth film on this substrate. The surface tension between the substrate and vapour is described by the 'effective tension' of the film which is a function of the film thickness $\gamma = \gamma_f(h)$ by analogy with a film on a solid substrate [14]:

$$\gamma_f(h) = \int_h^\infty h \frac{d\mu}{dh}\, dh + \gamma_{Ga/Bi} + \gamma_{Bi}. \tag{1}$$

$\gamma_{Ga/Bi}$ and γ_{Bi} are the tensions at the interfaces of two 'massive' liquids and μ is the chemical potential of the Bi film.

According to our idea a Bi film in the liquid state spreads over the Ga surface. The problem of spreading has been considered by de Gennes in the case of liquid films on a solid surface. In a one dimensional case the film thickness $\xi(x)$ depends only on a horizontal coordinate x. Then the film free energy is given by [8]

$$F = \int_{x_0}^{x_{max}} dx \left[-S + \frac{\gamma}{2}\left[\frac{d\xi}{dx}\right]^2 + P(\xi) + G(\xi) \right]. \tag{2}$$

S is the spreading coefficient which is the driving force for this process. $P(\xi)$ is the energy (per unit area) due to van der Waals forces and $G(\xi) = \rho g(\xi^2/2 + \xi L)$ is the hydrostatic surface energy and L is Lagrange's variational coefficient.

In the case of the spreading of a film on a *liquid* substrate an additional term stemming from the tension gradient along the surface should be introduced in the surface energy [9]. The surface tension gradient $\nabla \gamma_f$ as the main driving force of spreading (see [10]) substitutes for the spreading coefficient S in equation (2). The gradient of the tension arises from the difference in the film thickness along x (see figure 1). Bi film flows along $\nabla \gamma_f = (d\gamma_f/d\xi)(d\xi/dx)$ from the lower tension (film thickness ξ_0) to the higher tension (thickness ξ_1) where $\xi_1 < \xi_0$. Minimization of the modified equation (2) gives the first integral

$$\frac{\gamma}{2}\left[\frac{d\xi}{dx}\right]^2 = P(\xi) + G(\xi) - \nabla \gamma_f \xi(x). \tag{3}$$

To find an equilibrium film thickness h_0 we use the boundary condition in the centre of symmetry of the film $(d\xi/dx)/_{h_0} = 0$. Then we obtain a simple balance between hydrostatic pressure and van der Waals forces and the gradient of the tension

$$P(h_0) + G(h_0) = P(h_0) + \frac{3}{2}\rho g h_0^2 = h_0 \nabla \gamma_f = \frac{d\gamma_f}{dh}\frac{dh}{dx}. \tag{4}$$

Equation (3) together with the boundary condition in the form of equation (4) should be solved in order to find the film profile $\xi(x)$. Because the tension derivative $d\gamma_f/dx$ is essentially unknown to the present study we restrict our calculations to simple order of magnitude estimates of the equilibrium thickness h_0. For this we need the magnitude of $d\gamma_f/dx$ which can be estimated using a simple linear profile of the tension along the surface length L: $\nabla \gamma_f \sim \delta\gamma/L$.

We identify $\delta\gamma_f$ with the difference in tension between Ga covered with a macroscopic Bi film and Ga covered by a Bi monolayer. Using equation (1) we obtain

$$\delta\gamma_f = \int_h^\infty h\frac{\mathrm{d}\mu}{\mathrm{d}h}\,\mathrm{d}h + \gamma_{Bi} \tag{5}$$

where $\gamma_{Ga/Bi}$ is assumed to be essentially the same for both films: micro- and macroscopic. In the limit of $h \to \infty$ the integral in equation (5) tends to zero reflecting the fact that the van der Waals forces are not important for macroscopic thick films. Then $\delta\gamma_f$ is equal to the tension at the upper boundary of the Bi film, i.e. γ_{Bi} taken at some temperature ($T \sim 100\,^\circ\mathrm{C}$) much lower than the melting point of Bi. If we neglect the van der Waals term in equation (4) the tension $\delta\gamma$ can be compared to the gravitational term using the film thickness ($h_0 \sim 50\,\mu$) observable in the present experiment: $\delta\gamma \sim h_0\rho g L = 50 \times 10^{-4} \times 10 \times 10^3 \times 3 \sim 150$ erg cm^{-2}. This value agrees reasonably with the order of magnitude of the tension of liquid Bi especially if the tension of such a supercooled film is supposed to have an inversion in the $\gamma(T)$-behaviour far below the melting point. Our order of magnitude estimations of the equilibrium film thickness show that thick Bi films of the order of $50\,\mu$ can be obtained on a Ga surface in the course of spreading.

4. 'Classical' wetting in the Ga–Bi system

To obtain the light scattering spectra one should try to achieve shallow liquid Ga samples at room temperature. For this we used a glow discharge in argon as briefly described in [11]. Liquid Ga was dropped in a vacuum onto the Ti substrate and a voltage of about 4 kV was applied between the substrate and a needle-shaped electrode spaced at a distance of 5 cm apart. A glow discharge ignited at a pressure 10^{-2} Torr of argon. At this moment the anode–cathode voltage fell to 1 kV and the discharge current rose to about 1 mA integrally. It is enough to roll slightly the sample in order to obtain spreading of Ga over the container area. Ga spreads easily along the container wall until a ring of liquid is formed. Then the layer can be completed by shaking the sample. Layers prepared in such a way were usually between 1 and 2 mm thick and remained stable after switching off the field.

A theoretical explanation of this irreversible electric wetting is not obvious. There is an additional shear force acting at the liquid surface in an electric field which induces the liquid motion: $F_s = \epsilon_0 E_x E_z$, where ϵ_0 is the dielectric constant. Note that a reversion of the voltage polarity has no effect on F_s because the signs of both the surface charge density and the tangential field are changed. The tangential component E_x appears on the metal surface only when the current flows through the metal: $J_x = \sigma E_x$ (see figure 2). The spreading happens due to a decrease in the surface tension in the external electric field. This is not a well known electrocapillary effect but rather a reduction in the tension because of the momentum transfer at the surface subjected to a flux of Ar$^+$ in gas discharge. Mathematically the reduction in tension is given by $\delta\gamma = -j_s\delta u_s$ [12], where j_s is the density of momentum at the surface and u_s is the surface velocity appearing due to the spreading. The connection between j_s and u_s is given by a standard relation $j_s = \rho u_s$ [12] (ρ is the liquid density). In the present case the effect of the surface shear force leads to the velocity field inside the drop. Hence the reduction in the surface energy E_s is essentially equal to the kinetic energy of motion and is given by the integral $E_s = \rho \int_0^h \mathrm{d}z u^2$. The velocity at the liquid surface can be found from the balance between viscous and electric shear stresses which is not a simple task taking into account the non-uniform field and the charge density. However we may estimate the order of magnitude of the surface energy using a typical surface velocity $u_s \sim 1$ cm s^{-1} and a linear decay of u into the liquid depth δz of about 1 mm: $\delta E_s \sim \rho\delta z u^2/3 = 0.6$ erg cm^{-2}. The surface velocity
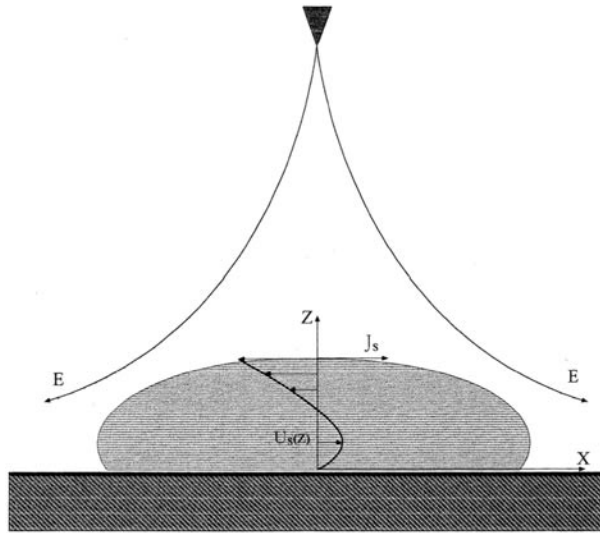
Figure 2. Wetting between a Ga drop and a metal substrate in glow discharge. The electric field together with the surface current produce the surface force which drives the spreading of the liquid drop. The velocity distribution inside the drop is also shown.

of 10 cm s^{-1} leads to $\delta E = 300$ erg cm^{-2} that shows the correct order of magnitude of the influence of the electric field on spreading.

Due to the reduction in the layer thickness capillary wave spectra from the liquid sample can be collected. Capillary waves are driven by the surface tension at the interface between Ga substrate and a film of Bi. The temperature derivative of the tension is given by the standard Gibbs relation where the surface concentration $\Gamma_s > 0$ has only conditional meaning for films thicker than a monolayer; it is replaced by the film thickness h [14]:

$$\frac{\partial \gamma_{Ga/Bi}}{\partial T} = \frac{\partial}{\partial T}(-\Delta S \, \mathrm{d}T - \Gamma_s \, \mathrm{d}\mu_s) = -\Delta S - \frac{h\rho}{M}\frac{\partial \mu_s}{\partial T} \tag{6}$$

where ρ and M are the density and the molecular weight of Bi respectively. One can see that a sign of $\partial \gamma_{Ga/Bi}/\partial T$ depends on the interplay between the entropy term and the temperature derivative of the film chemical potential μ_s. The surface entropy is mainly given by the ratio of two mean frequencies of thermal excitations [13]:

$$\Delta S = n_s k_B \ln \frac{\bar{\omega}_p}{\bar{\omega}_c} \tag{7}$$

where n_s is the number of atoms per unit area, $\bar{\omega}$ is the mean frequency of capillary waves, defined as $\bar{\omega}_c = (1/n_s) \int_0^{q_{max}} \mathrm{d}q\omega(q)2\pi q$ and $\bar{\omega}_p$ is the mean frequency of bulk phonons. According to Frenkel's analysis this part of the surface excess entropy is positive because $\bar{\omega}_p > \bar{\omega}_c$. However as we have shown in [15] the surface entropy can be split into different parts: the first is due to capillary waves. The second part is an orientational one and is determined by surface layering and anisotropic pressure in the surface layer. Under certain circumstances this part may become negative setting up a positive slope of the temperature derivative of surface tension at pure Ga. Having in mind that surface layering remains on surfaces of Ga–Bi alloys we cannot exclude its influence on the negativity of ΔS at least for very diluted alloys [1, 4]. The chemical potential of the film can vary in different ways depending on the behaviour of the function $\mu(h)$ [14]. A condition for mechanical stability of
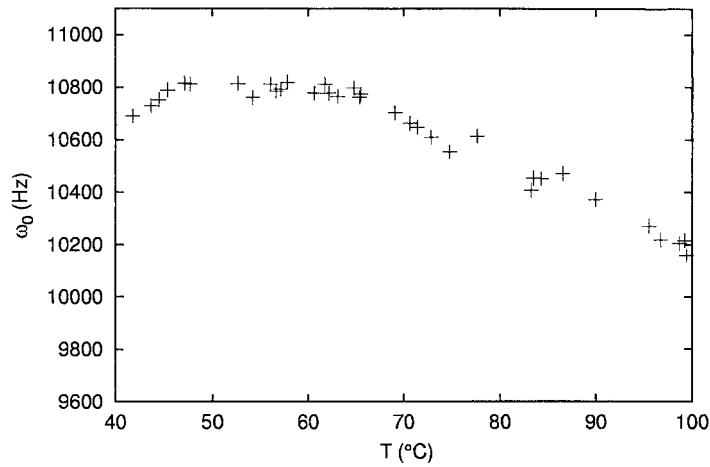
Figure 3. Temperature variations of peak frequencies of capillary waves at the gallium–bismuth interface. The errors in ω_0 are less than the size of the data points.

a film is $\partial\mu_s/\partial h > 0$ [14]. If we expect the thickness h to grow on heating ($\partial h/\partial T > 0$) then $\partial\mu_s/\partial T > 0$.

The light scattering spectra were collected in the T-interval from 40 to 120 °C. Each spectrum was fitted with a theoretical spectral form which is the exact replica of the autocorrelation function transformed to the frequency domain [7]

$$S(\omega) = A\Re[\exp(-(i\Gamma/\beta + (\omega - \omega_0)/\beta)^2)\,\mathrm{erfc}(-i[i\Gamma/\beta + (\omega - \omega_0)/\beta])] + B. \tag{8}$$

Thus, five properties were extracted from the fit of experimental spectra: frequency ω_0, damping constant Γ, instrumental width β, amplitude A and background B. In the present context we concentrate only on peak frequencies ω_0. To first order, the roots of the dispersion relation describing the propagation of a capillary wave with a particular wavenumber q are [5]

$$\omega_0 = \sqrt{\gamma q^3/\rho} \tag{9}$$
$$\Gamma = 2\eta q^2/\rho \tag{10}$$

where γ, η and ρ are the surface tension, bulk viscosity and density, respectively. Also the presence of a surface film modifies the dispersion equation in such a way that the damping constants Γ strongly deviate from the solution given by equation (10). However the peak frequencies are affected by the surface film by not more than 10% as shown in [5]. Therefore equation (9) serves as a basis for evaluation of the apparent tension which reflects some error introduced by the usage of equation (9). Using the peak frequency $\omega_0 = 10.7\,\mathrm{kHz}$ at $T = 40\,°\mathrm{C}$ and $q = 366\,\mathrm{cm}^{-1}$ the surface tension inferred from equation (9) is $\gamma = 562\,\mathrm{mN\,m}^{-1}$.

The light scattering data do not show a decreasing dependence $\omega_0(T)$ in the whole T-interval (figure 3). This indicates that $\gamma(T)$ is not a monotonic function and the surface tension increases on heating from 40 to 60 °C. In principle, both terms in equation (6) might change the sign of the tension derivative: the entropy term ΔS can be negative or the film thickness may decrease with T setting up $\partial\mu_s/\partial T < 0$. The second possibility reported for very dilute alloys of gallium–lead [3] is very likely to be the case in our system which is thermodynamically similar to Ga–Pb. If the decrease in the thickness of Bi is supposed to have the main influence on the T-dependence of the tension this implies a *dewetting* transition in the interval from 40 to 60 °C. A similar tendency, namely the reduction in the surface concentration

on heating from 25 to 77 °C was reported in the study of alloys of the composition 0.18 at.% Bi in Ga [4]. A theoretical explanation is not simple because both surface and bulk concentration have been changed with temperature. Above $T \sim 70\,°C$ $\gamma(T)$ switches to a more usual decreasing function that can be explained by the influence of both terms in equation (6) (see above).

After heating to 100 °C the sample was cooled for one night to room temperature in order to monitor possible surface changes. Even at T of about 31 °C no solid Bi film was detected. However the positions of the diffraction maxima (and q) were shifted indicating that the reflection angle has also been changed due to the changes in contact angle between liquid sample and container wall. These changes were irreversible: upon heating up to 100 °C the positions of the diffraction maxima remained unaffected. Peak frequency collected from 100 to 160 °C displayed a steadily decreasing tendency with nearly the same slope as a descendent tail of the data in figure 3. We emphasize that the shape of the diffraction maxima was altered on heating above 120 °C because the surface curvature was T-dependent. In such a situation it was nearly impossible to keep q constant which is vitally essential for the data evaluation. Therefore we present no data and report only the results of visual observations in the T-interval from 120 to 220 °C.

To follow the evolution of the Bi film the sample temperature was raised and decreased gradually. Only after heating above 190 °C did Bi accumulate on the surface such that a solid island was visible on cooling the sample. At T of about 220 °C a convective motion was visible at the liquid surface. This motion was laminar with a much lower velocity than in previous experiments without wetting between Ga and container wall. This is Marangoni convection induced by the non-uniform distribution of the Bi film along the surface. This point is supported by visual observations on a solidified sample whose surface was only partially covered by Bi film having a typical yellow colour. Thus we cannot exclude the influence of the flow on the transfer of Bi through defects at the Ga–container contact line.

In sum, two experiments on the same system show two principally different results. The first clearly indicates that a macroscopic Bi film is formed on Ga at some temperature far away from the monotectic point in the bulk phase diagram. The second shows that this film appears at a much higher temperature very close to the coexistence of two liquids. This is a clear demonstration of the idea about the driving force of wetting transition. According to our suggestions this transition happens in the course of the spreading of Bi over the surface of liquid Ga. When Ga perfectly wets the container wall the Bi film cannot flow along the wall and the spreading is greatly suppressed. In such a situation it is likely that the appearance of a Bi film is driven by critical fluctuations at some T close to the monotectic point of the Ga–Bi system.

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