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Spreading of a mercury droplet on thin gold films

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Abstract

The spreading of a small mercury droplet (150 μm) on thin gold films is studied, using an optical microscope enhanced with a differential interference contrast system. The growing interfaces are analyzed in order to determine the roughness (α) and growth (β) exponents. For gold film thickness of 1500 Å we find that $\alpha = 0.88 \pm 0.03$ and $\beta = 0.76 \pm 0.03$, while for gold thickness of 3000 Å, $\alpha = 0.96 \pm 0.04$ and $\beta = 1.00 \pm 0.04$. Both sets of exponents satisfy the scaling relation $\alpha + \alpha/\beta = 2$. In both systems the roughness exponent α crosses over to a value close to 0.5 in the final stages of the experiment and for relatively long length scales (order of a few microns).

Keywords: Thin films; Surface growth; Diffusion; Reaction

Spreading of liquids on solids and wetting of solid surfaces contain rich physical, chemical, and statistical phenomena [1]. In earlier works [2,3], we studied the dynamics and geometry of the spreading process of a mercury droplet on a thin *silver* film using an optical microscope equipped with a differential interference contrast (DIC) system (Fig. 1). The Ag films were thermally deposited on microscope slides in a vacuum chamber. The Hg front propagation was recorded by a video camera. The images were analyzed in order to determine the geometrical and dynamical properties of the front.

Modern statistical methods [4–7] of analyzing interface lines use two exponents to characterize the interface and associate it with a specific universality class. The

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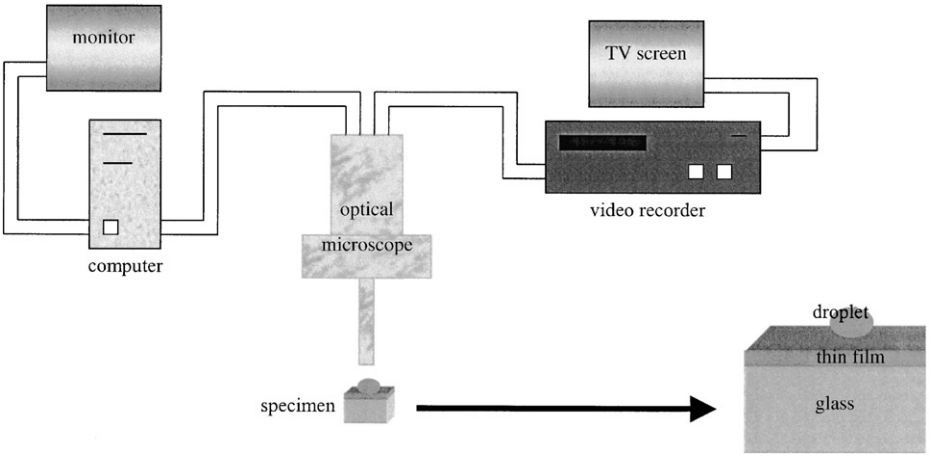

Fig. 1. A schematic view of the experimental system.

Fig. 2. The function $h(x,t)$ for describing the interface characteristics, representing a top view of the experiment. The width W satisfies Eqs. (1) and (2).

roughness exponent α and the growth exponent β are based on a function $h(x,t)$ associated with the top view of the front line (Fig. 2). A width function W is then defined as

$$W^2(L,t) = \langle h(x,t)^2 \rangle - \langle h(x,t) \rangle^2 \,, \tag{1}$$

where L varies from the smallest length scale to the system size L_0. The width W satisfies [5]

$$W \sim \begin{cases} t^{\beta}, & t \ll t_0 \,, \\ L^{\alpha}, & t \gg t_0 \,, \end{cases} \tag{2}$$

where $t_0 \approx L^{\alpha/\beta}$.

In the Ag–Hg system [2,3], we have studied the spreading of small droplets (50–200 μm) of Hg on thin Ag films. For Ag film thickness of 500–2000 Å we found that $\alpha = 0.66 \pm 0.03$ and $\beta = 0.46 \pm 0.02$, while for film thickness of 0.1 mm, $\alpha = 0.77 \pm 0.04$ and $\beta = 0.60 \pm 0.02$. Both sets of exponents satisfy the scaling relation $\alpha + \alpha/\beta = 2$. In both cases, the roughness exponent α has been shown to cross over to about 0.5 in the final stages of the experiment and for relatively long length scales (order of a few microns).

In this work, we study the dynamics and geometry of the interface in a similar system, replacing the thin silver films by thin *gold* films. This is done since it was shown [2,3,8–11] that the roughness of the substrate on which the droplet is running plays an important role in the characterization of the moving interface. The smoother the substrate, the smoother is its interface with the droplet. In general, Au surfaces are found to be much smoother than Ag surfaces [12–15]. Thus, for the Au–Hg system one expects different roughness and growth exponents compared to the Ag–Hg system.

In addition, the chemical reaction between the droplet and the substrate is an important mechanism in the process, in particular at its late stages. The reaction Ag–Hg differs from Au–Hg both in the number of possible alloys and in the atomic percent in a specific inter-metallic compound [16]. Possible reactions in the silver–mercury system are: $4Ag + 3Hg \rightarrow Ag_4Hg_3$ and $3Ag + 4Hg \rightarrow Ag_3Hg_4$ while for the gold–mercury system one has $5Au + Hg \rightarrow Au_5Hg$, $3Au + Hg \rightarrow Au_3Hg$, $2Au + Hg \rightarrow Au_2Hg$ and $Au + 2Hg \rightarrow AuHg_2$. Therefore, grains of the new alloys that grow on the interface between the materials differ from each other in their structure and may yield different roughness and growth exponents.

The Au films, in two different thicknesses, 1500 Å and 3000 Å, were thermally deposited on microscope slides in a vacuum chamber, in order to create well-controlled clean smooth surfaces along relatively large distances. The Hg droplet (150 μm in diameter) was placed on the gold substrate a few minutes after the evaporation. The experiment was performed at room conditions. The spreading process was monitored, recorded and analyzed using the method described in Ref. [2].

Using Eqs. (1) and (2) we have analyzed the Au–Hg interfaces and found $\alpha = 0.88 \pm 0.03$ (Fig. 3a) and $\beta = 0.76 \pm 0.03$ (Fig. 3b), in the thin (1500 Å) Au film systems, averaged over five experiments. Fig. 3 represents the data of a single experiment. The two exponents satisfy the well-known scaling relation for isotropic systems $\alpha + \alpha/\beta = 2$ [7], within the error bars. When the spreading process completely stopped, we calculated α again and found that $\alpha \approx 0.85$ for short length scales ($L_c < 7$ μm), and $\alpha \approx 0.49$ for long length scales ($L_c > 7$ μm) (Fig. 3c).

For thicker gold substrates (3000 Å), the exponents were found to be $\alpha = 0.96 \pm 0.04$ (Fig. 4a) and $\beta = 1.00 \pm 0.04$ (Fig. 4b), averaged over five experiments as well. These exponents, although different from the thinner film exponents, still obey the scaling relation $\alpha + \alpha/\beta = 2$. When the spreading process stopped, we calculated α again and found $\alpha \approx 0.86$ for short length scales ($L_c < 14$ μm), and $\alpha \approx 0.48$ for long length scales ($L_c > 14$ μm) (Fig. 4c).

The high value of α (close to 1) during the process indicates strong correlation between the points on the interface. This reflects the high surface tension along the perimeter of the propagating droplet and a significant wetting process. The smaller value of $\alpha \approx 0.88$ in the thinner system (1500 Å) compared to that in the thicker one (3000 Å), $\alpha \approx 0.96$, is probably due to the underlying glass that disturbs the wetting by touching the droplet and creating dewetting forces along some of the points of the interface. Consequently, the interface becomes rougher and the roughness exponent is decreasing. It can also be compared to the Ag–Hg system in which the *in-situ* roughness exponents were smaller. This is because the silver has a much rougher surface that breaks the symmetry of spreading.

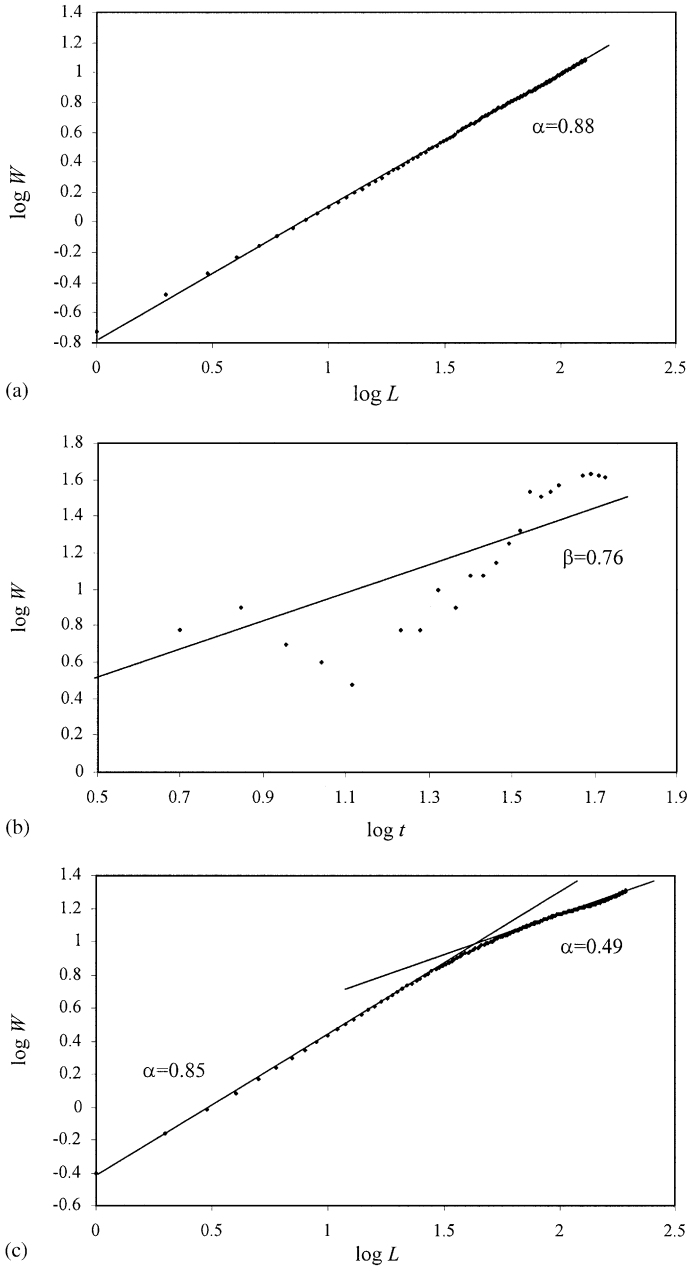

Fig. 3. (a) The roughness exponent α, and (b) the growth exponent β, in the thinner system (1500 Å), during the experiment; (c) the crossover behavior of the roughness exponent at the end of the process.

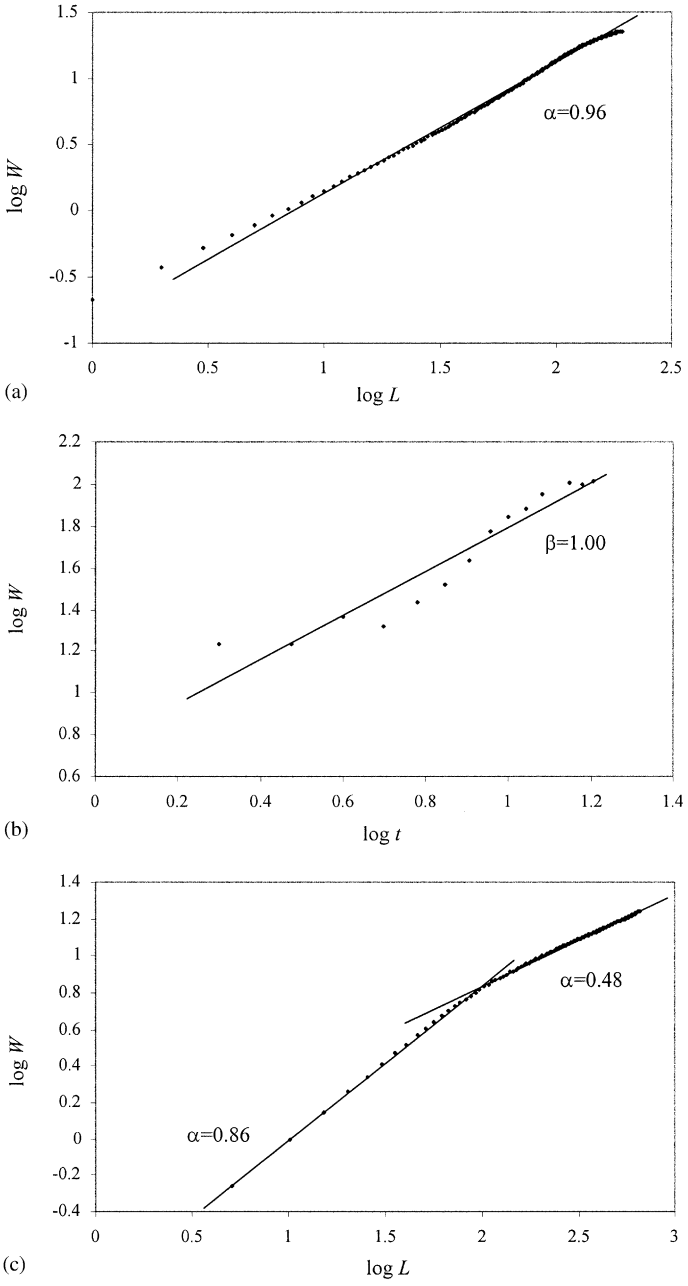

Fig. 4. (a) The roughness exponent α, and (b) the growth exponent β, in the thicker system (3000 Å), during the experiment; (c) the crossover behavior of the roughness exponent at the end of the process. The crossover takes place at a larger distance compared to the thinner system shown in Fig. 3.

Table 1
Summary of roughness and growth exponents

	During the process	End of process
Thin film 1500 Å	$\alpha = 0.88 \pm 0.03$ $\beta = 0.76 \pm 0.03$	$\alpha_{L<7\ \mu m} \sim 0.85$ $\alpha_{L>7\ \mu m} \sim 0.49$
Thick film 3000 Å	$\alpha = 0.96 \pm 0.04$ $\beta = 1.00 \pm 0.04$	$\alpha_{L<14\ \mu m} \sim 0.86$ $\alpha_{L>14\ \mu m} \sim 0.48$

When the spreading process stops, surface tension forces are much smaller than the chemical reaction between the Hg and the Au substrate and a rougher interface is formed. The short-scale correlation of points along the interface is due to the reaction that grows the new nuclei of the inter-metallic compound in a certain structure. For long scales, there is no correlation between points on the interface since they do not belong to the same nucleus and the roughness exponent α decreases to about 0.5 [17]. In the thicker film (3000 Å), the correlation length is longer ($L_c \approx 14\ \mu m$) than in the thinner system (1500 Å) ($L_c \approx 7\ \mu m$), because the bigger gold bulk causes bigger grains of the new inter-metallic compounds.

To summarize, we presented data on the geometry and dynamics of a gold–mercury reaction interface in a spreading droplet experiment. The roughness and growth exponents are summarized in Table 1. The crossover behavior of the roughness exponent sheds light on different mechanisms and correlation lengths in the process.

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References

[1] P.G. de Gennes, Rev. Mod. Phys. 57 (1985) 828.
[2] A. Be'er, Y. Lereah, H. Taitelbaum, Physica A 285 (2000) 156.
[3] A. Be'er, Y. Lereah, I. Hecht, H. Taitelbaum, Physica A 302 (2001) 297.
[4] A. Bunde, S. Havlin, Fractals and Disordered Systems, 2nd Edition, Springer, Berlin, 1996.
[5] A. Bunde, S. Havlin, Fractals in Science, 2nd Edition, Springer, Berlin, 1994.
[6] P. Meakin, Fractals, Scaling, and Growth Far from Equilibrium, Cambridge University Press, Cambridge, 1998.
[7] T. Vicsek, Fractal Growth Phenomena, 2nd Edition, World Scientific, Singapore, 1992.
[8] H.G.E. Hentschel, F. Family, Phys. Rev. Lett. 66 (1991) 1982.
[9] D.A. Kessler, H. Levine, Y. Tu, Phys. Rev. A 43 (1991) 4551.
[10] O. Zik, T. Kustanovich, E. Moss, Z. Olami, Phys. Rev. E 58 (1998) 689.
[11] M.H. Jensen, I. Procaccia, J. Phys. II1 (1991) 1139.
[12] R. Chiarello, V. Panella, J. Krim, C. Thompson, Phys. Rev. Lett. 67 (1991) 3408.
[13] J. Krim, I. Heyvaert, C. Van Haesendock, Y. Bruynseraede, Phys. Rev. Lett. 70 (1993) 57.
[14] M. Kardar, J.O. Indekeu, Phys. Rev. Lett. 65 (1990) 662.
[15] P. Pfeifer, M.W. Cole, J. Krim, Phys. Rev. Lett. 65 (1990) 663.
[16] M. Hansen, Constitution of Binary Alloys, 2nd Edition, McGraw-Hill, New York, 1958.
[17] M. Kardar, G. Parisi, Y.C. Zhang, Phys. Rev. Lett. 56 (1986) 889.