# XVIII. Surface-Induced Disorder and Surface Melting

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## XVIII.1. Introduction and Outline

Consider a uniaxial antiferromagnet on a simple cubic lattice in an external magnetic field. The lattice can be divided into two sublattices such that, at low temperature $T$, all spins within one sublattice point in the same direction. In fact, there are two distinct ordered states, at a given $T$, which can be transformed into one another by interchanging the two sublattices. With increasing $T$, the magnet becomes less ordered up to a transition temperature $T = T_\star$ at which it undergoes a phase transition to a disordered state. I will be concerned here with the case where this transition is *first order*. Thus, if we look into the *bulk* of such an antiferromagnet, we will see *discontinuous* change from the ordered to the disordered state.

Now, assume that we look instead at the *surface* of such a magnet. Naively, one would expect to see again a discontinuous change in the order. However, theoretical models typically lead to a rather different behavior since they predict a *continuous* decrease of the surface order for a wide range of microscopic coupling parameters. In these cases, any surface order parameter, $M_1$, is found to behave as

$$M_1 \sim (T_\star - T)^{\beta_1} \text{ with } \beta_1 > 0. \qquad (XVIII.1)$$

The basic physical mechanism for the continuous surface behavior is as follows. Let us decompose the semi–infinite system into a 2–dimensional surface region and a 3–dimensional bulk domain. If there were no coupling between those two systems, the surface would become disordered at its own transition temperature, $T = T_\star^{(s)}$. If the couplings within the surface are comparable to those within the bulk, mean field theory leads to the rough estimate $T_\star^s \cong 2T_\star/3$. Thus for the temperature interval $T_\star^{(s)} < T < T_\star$, the surface would be disordered while the bulk is ordered. Of course, this picture is too crude since the surface *is* coupled to the bulk. On the one hand, this coupling leads to the residual order within the surface region as given by (XVIII.1) as long as $T < T\star$. On the other hand, it implies that the disordering, which is initiated at the surface, spreads into the interior of the crystal. As a result, a whole layer or film of the (nearly) disordered phase intrudes between the surface and the ordered bulk. This process has been termed surface–induced disorder (SID) [XVIII.1].

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It is obvious from the picture just described that SID is *not* restricted to first–order transitions in antiferromagnets. In fact, such a behavior was first found theoretically for the (100) surface of the Potts model on a simple cubic lattice [XVIII.2,XVIII.3]. In this case one has $q \geq 3$ ordered states which undergo a first–order transition to the disordered phase. More generally, SID can occur for any first–order phase transition with spontaneously broken symmetry.

One example for SID which has been experimentally studied in some detail is the (100) surface of $Cu_3Au$. This binary alloy undergoes an order–disorder transition at $T_* = 663\,K$ which is strongly first–order in the bulk. In contrast, some LEED data indicated that the surface order vanishes in a continuous fashion [XVIII.4], which led us to suggest that this system provides an example for SID [XVIII.5]. During the last five years, more experimental data have been obtained by LEED [XVIII.6], spin–polarized LEED [XVIII.7], and X–ray diffraction under total external reflection [XVIII.8], which are all consistent with this view.

So far, I have discussed phase transformations within a crystal which do not affect the translational order of the crystal lattice. Now, let us consider the most drastic change in the translational order of the crystal, i.e., melting. The idea that melting often starts at the crystal surface has a long history [XVIII.9]. However, it has been realized only recently that the process of surface melting can lead to surface critical behavior as in (XVIII.1) [XVIII.1]. Much of the recent experimental work on surface melting has focused on Pb, for which it was observed by ion scattering [XVIII.10,XVIII.11] and LEED [XVIII.12]. In addition, surface melting has been recently studied for Ar [XVIII.13], $O_2$ [XVIII.14], methane [XVIII.15], Ne [XVIII.16] and Ge [XVIII.17].

In the remainder of this paper, I will give a brief review on surface–induced disorder and surface melting, which emphasizes the basic theoretical concepts. First, the free energy of the disordered surface layer is discussed in Sec. 2. This free energy contains the direct interaction of the two interfaces bounding the layer. The form of this interaction depends on the intermolecular forces (Sec. 3). As a consequence, the critical exponents $\beta_1$ which govern the surface order parameters as in (XVIII.1) are, in general, non–universal and depend on microscopic parameters (Sec. 4). Finally, I will give a brief outlook on open questions (Sec. 5).

## XVIII.2. Free Energy of the Disordered Surface Layer

Consider a disordered surface layer of thickness $\ell$, and let us estimate its free energy per unit area, $V(\ell)$. First of all, one must note that the interface between the crystal and the vapor (or 'vacuum ') surrounding it now consists of two interfaces: (i) the (vd) interface between the disordered surface phase and the vapor, and (ii) the (do) interface between the disordered surface layer and the ordered bulk state of the crystal. Thus, the free energy $V(\ell)$ may also be regarded as the *effective interaction* between two interfaces.

The leading term of $V(\ell)$ for large $\ell$ arises from the fact that the disordered phase is metastable rather than stable for $T < T_\star$. Close to $T_\star$, the bulk free energy per unit volume, $f_{vo}$, of the ordered crystalline phase, (which coexists with the vapor) behaves as $f_{vo} \approx f^\star - S^\star_{vo}(T - T_\star)$. Likewise, the bulk free energy, $\hat{f}_d$, of the metastable disordered phase is given by $\hat{f}_d \approx f^\star - S^\star_d(T - T_\star)$. Then the difference in the bulk free energies $f_{vo}$ and $\hat{f}_d$ leads to $V(\ell) \approx \Delta S(T - T_\star)$ with $\Delta S \equiv S^\star_d - S^\star_{vo}$ for a layer of thickness $\ell$.

At $T = T_\star$, the disordered phase coexists with the ordered phase (and with the vapor), and the leading term of $V(\ell)$ is given by the sum, $\Sigma_{vd} + \Sigma_{do}$, of the interfacial tensions of the two interfaces. The next–to–leading terms, which reflect the nature of the underlying intermolecular forces, will be referred to as the *direct interaction*, $V_{DI}(\ell)$, between the (vd) and the (do) interfaces. Thus, the free energy per unit area of the disordered surface layer has the generic form

$$V(\ell) \approx \Delta S(T_\star - T)\ell + \Sigma_{vd} + \Sigma_{do} + V_{DI}(\ell) \qquad (XVIII.2)$$

for large $\ell$ with $V_{DI}(\ell = \infty) = 0$.

In mean–field theory, the equilibrium value of $\ell$ follows from [XVIII.1] $\partial V/\partial \ell = 0$ or

$$\Delta S(T_\star - T) = -\partial V_{DI}(\ell)/\partial \ell. \qquad (XVIII.3)$$

Two cases must be distinguished depending on the sign of $V_{DI}(\ell)$ for large $\ell$. First, assume that $V_{DI}(\ell)$ and, thus, $-\partial V_{DI}/\partial \ell$ is positive for large $\ell$. In this case, (XVIII.3) implies that the layer becomes thicker and thicker as $T_\star$ is approached from below [XVIII.18], and the disordered surface layer becomes more and more similar to the disordered bulk phase. This is the process of *complete* surface–induced disorder (or surface melting) with $\Sigma_{vo} = \Sigma_{vd} + \Sigma_{do}$ at $T = T_\star$. On the other hand, one may have $V_{DI}(\ell) < 0$ and thus $-\partial V_{DI}/\partial \ell < 0$ for large $\ell$. In the latter case, either no disordered layer appers in the surface region or the thickness of this layer remains finite as $T_\star$ is attained [XVIII.18]. This is the case of *incomplete* surface–induced disorder (or surface melting) with $\Sigma_{vo} < \Sigma_{vd} + \Sigma_{do}$ at $T = T_\star$.

In a fluid context, the two relations $\Sigma_{\alpha\gamma} = \Sigma_{\alpha\beta} + \Sigma_{\beta\gamma}$ and $\Sigma_{\alpha\gamma} < \Sigma_{\alpha\beta} + \Sigma_{\beta\gamma}$ for the interfacial tensions of three coexisting phases $\alpha$, $\beta$ and $\gamma$ are well–known and correspond to complete and incomplete wetting, respectively [XVIII.19]. Thus, surface–induced disorder (or surface melting) and wetting belong to the same class of interfacial phase transitions [XVIII.20,XVIII.21,XVIII.22]. However, in contrast to fluid systems, the interfacial tensions $\Sigma_{vo}$, $\Sigma_{vd}$, and $\Sigma_{do}$ for the systems considered here depend on the *orientation* of the crystal surface. Therefore, the relation $\Sigma_{vo} = \Sigma_{vd} + \Sigma_{do}$ may hold for some surface orientations, but not for others, and a macroscopic crystal can exhibit both ordered and disordered facets (or rounded portions of its surface) at the same time. This *anisotropy* of the surface behavior has been observed for the surface melting of Pb [XVIII.11].

## XVIII.3. Direct Interaction of Interfaces Bounding the Surface Layer

As mentioned above, the term $V_{DI}(\ell)$ in (XVIII.2) represents the direct interaction between the two interfaces bounding the surface layer, and arises from the microscopic forces between atoms and molecules. First, let us assume that the short-range part of these forces is strong, and let us ignore possible contributions to $V_{DI}(\ell)$ from their long-range part. Then, $V_{DI}(\ell)$ decays *exponentially* for large $\ell$. The corresponding decay length can arise from an *order parameter* (OP) density or from a *non-ordering* (NO) density.

In general, a macroscopic system can be described by several OP and NO densities. By definition, all OP densities vanish and all NO densities have a finite value when the system is in disordered high-$T$ state. For example, the Potts model on a simple cubic lattice with $q = 3, 4, \ldots$ states is described by $(q-1)$ OP densities. Likewise, an antiferromagnet on a simple cubic lattice is characterized by one OP and one NO density which are given by the difference and the sum of the sublattice magnetizations, respectively.

For a system governed by short-range forces, all densities decay exponentially $\sim \exp(-z/a)$ with the distance $z$ from an interface but the size of the decay length, $a$, depends on the density considered. Let $\xi_d$ denote the *largest* decay length of all OP densities in the disordered phase, and let $\kappa_d$ be the largest decay length of all NO densities [XVIII.23]. Then, two cases must be distinguished. For $\xi_d/2 > \kappa_d$, the OP density governed by $\xi_d$ dominates and the direct interaction decays as [XVIII.1,XVIII.24]

$$V_{DI} \approx c_1 \, \Sigma_{do} \, exp[-2\ell/\xi_d] \qquad (\xi_d/2 > \kappa_d) \qquad (XVIII.4)$$

for large $\ell$ where $c_1 \sim O(1)$ is a numerical coefficient.

It is interesting to note that the decay length for $V_{DI}(\ell)$ is $\xi_d/2$ rather than $\xi_d$. This property is a consequence of the fact that the systems considered here have several ordered phases for $T < T_*$ (which are related by a discrete symmetry) and, thus, have a vanishing field conjugate to the OP densities.

On the other hand, for $\kappa_d > \xi_d/2$, the NO density governed by $\kappa_d$ dominates and the direct interaction has the asymptotic behavior [XVIII.25]

$$V_{DI}(\ell) \approx \bar{c}_1 \, \Sigma_{do} \, exp[-\ell/\kappa_d] \qquad (\kappa_d > \xi_d/2) \qquad (XVIII.5)$$

for large $\ell$. Usually, the numerical coefficient $\bar{c}_1$ is again $\sim O(1)$ but, in some cases, $\bar{c}_1$ is exceptionally small. Then, one must include the next-order term which is given by $exp[-2\ell/\kappa_d]$, $exp[-2\ell/\xi_d]$ or $exp[-\ell/\kappa'_d]$ where $\kappa'_d$ is the second largest decay length of the NO densities.

Next, let us consider the contribution to $V_{DI}(\ell)$ which arises from the long-range part of the intermolecular forces. These contributions lead to a direct interaction,

$$V_{DI}(\ell) \approx -W/\ell^{r} \text{ for large } \ell, \qquad (XVIII.6)$$

which decays as a power law. Indeed, all atoms or molecules interact via long-range van der Waals forces. These forces give a direct interaction $V_{DI}(\ell) \sim 1/\ell^{r}$ for large $\ell$ with $r = 2$ and $r = 3$ for nonretarded and retarded forces respectively in 3-dimensional systems [XVIII.26].

## XVIII.4. Non-Universal Behavior of Surface Order Parameters

In a semi-infinite system bounded by a planar surface, the OP densities depend on the distance $z$ from the surface $z = 0$. Now, let $m^{(j)}(z)$ with $j = 1, 2, \ldots$ denote the different OP profiles. In order to simplify the discussion, I will assume in the following that these profiles *decay exponentially* with $z$: $m^{(j)}(z) \sim exp(-z/\xi_d^{(j)})$ where the decay lengths $\xi_d^{(j)}$ are ordered according to their size, i.e. $\xi_d^{(j)} \geq \xi_d^{(j+1)}$ with $\xi_d^{(1)} \equiv \xi_d$.

Within mean-field theory, the equilibrium value of the layer thickness, $\ell$, follows from $(T_{*} - T) \sim -\partial V_{DI}/\partial \ell$ as in (XVIII.3). Then, the mean-field behavior of the surface OPs, $m^{(j)}(z = 0)$, is given by

$$M_1^{(j)} \equiv m^{(j)}(z = 0) \sim exp[-\ell/\xi_d^{(j)}]. \qquad (XVIII.7)$$

For systems governed by short-range forces, the direct interaction $V_{DI}$ has the form as given by (XVIII.4) or (XVIII.5). First assume that $\xi_d/2 > \kappa_d$, i.e. that the dominant length scale belongs to an OP density. Then (XVIII.4), (XVIII.3), and (XVIII.7) imply the mean-field critical behavior

$$\ell \approx \frac{1}{2}\, \xi_d \, \ln[T_{SC}/(T_{*} - T)] \qquad (XVIII.8)$$

and

$$M_1^{(j)} \sim (T_{*} - T)^{\beta_1} \text{ with } \beta_1 = \xi_d/2\xi_d^{(j)}. \qquad (XVIII.9)$$

as $T_{*}$ is approached from below. Thus, the dominant OP density vanishes as $M_1^{(1)} \sim (T_{*} - T)^{\beta_1}$ with the universal critical exponent $\beta_1 = 1.2$ [XVIII.3].

Next, consider the case for short-range forces with $\kappa_d > \xi_d/2$ where $\kappa_d$ is again the decay length of the dominant NO density. Then, $V_{DI}(\ell)$ is given by (XVIII.5) which leads to

$$\ell \approx \kappa_d \ln[T_{SC}/(T_{*} - T)] \qquad (XVIII.10)$$

and

$$M^{(j)} \sim (T_* - T)^{\beta_1} \text{ with } \beta_1 = \kappa_d/\xi^{(j)} \qquad (XVIII.11)$$

for small $(T_* - T)$. Thus, the dominant OP density vanishes at the surface as $M_1^{(1)} \sim (T_* - T)^{\beta_1}$ with the parameter–dependent critical exponent $\beta_1 = \kappa_d/\xi_d$ [XVIII.25].

In general, the form of the direct interaction $V_{DI}(\ell)$ depends on the surface orientation. Therefore, the critical exponent $\beta_1$ as given by (XVIII.11) or (XVIII.13) will also depend on this orientation. This has been shown explicitly for a spin (or lattice gas) model which describes a binary alloy on a fcc lattice. In this case, the system is described by one OP density and one NO density which are given by the Bragg–Williams OP and the composition, respectively [XVIII.27,XVIII.25]. Within mean–field theory, the OP is governed by $\beta_1 = 1/2$ for the (111) and the (110) surfaces of the fcc lattice, and by $\beta_1 \cong 2.2 - 2.8$ for the (100) surface where, in the latter case, the precise value depends on the coupling parameters [XVIII.25].

The critical behavior as given by (XVIII.11) has also been obtained within a phenomenological model for surface melting [XVIII.28]. Quite generally, the atomic density of a semi–infinite crystal can be parametrized by the reciprocal lattice vectors, $\vec{Q}''$, of the 2–dimensional lattice parallel to the surface. Within the model of Ref. XVIII.28, $\tilde{M}(\vec{Q}'' = 0, z)$ is treated as a NO density while all $\tilde{M}(\vec{Q}'', z)$ with $|\vec{Q}''| > 0$ are treated as OP densities [XVIII.29]. One then finds that the surface OPs vanish as in (XVIII.11) with

$$\beta_1(\vec{Q}''_2) > \beta_1(\vec{Q}''_1) \text{ for } |\vec{Q}''_2| > |\vec{Q}''_1|. \qquad (XVIII.12)$$

Thus, the OP components with *larger* $|\vec{Q}''|$ are predicted to vanish *faster* as $T_*$ is approached. This is in qualitative agreement with recent LEED experiments on Pb [XVIII.12], and is expected to hold in more detailed models of surface melting.

Finally, consider the case of long–range forces and $V_{DI}(\ell) \sim 1/\ell^r$ as in (XVIII.6). Such an interaction with $r = 2$ (arising from non–retarded van der Waals forces) should apply to surface melting as soon as the disordered layer gets sufficiently thick [XVIII.21,XVIII.16,XVIII.30]. Then, the disordered surface layer thickens as [XVIII.31]

$$\ell \sim 1/(T_* - T)^{\psi} \text{ with } \psi = -\beta_s = 1/(1 + \mathrm{r}), \qquad (XVIII.13)$$

and the surface OPs vanish as [XVIII.31–33,XVIII.28]

$$M^{(j)} \sim exp[-c^{(j)}/(T_* - T)^{\psi}] \qquad (XVIII.14)$$

provided the OP profiles decay exponentially as assumed.

## XVIII.5. Summary and Outlook

In summary, the surface of a crystal may initiate the disordering process near a first–order phase transition well below the transition temperature $T = T_*$. Then, the surface exhibits critical behavior and the surface OPs vanish as in (XVIII.1) with characteristic critical exponents $\beta_1$. In general, these exponents are non–universal and reflect the relative size of microscopic length scales [see (XVIII.9), (XVIII.11), and (XVIII.12)].

In some systems, the surface favors the ordered state rather than the disordered one. Such surface–induced order [XVIII.1] or surface freezing has been observed experimentally at first–order phase transitions between crystalline phases [XVIII.34]. In these cases, the surface OPs do not vanish, and the thickening of the ordered surface layer is governed by the direct interactions (XVIII.5) or (XVIII.6) which leads to the growth laws (XVIII.10) or (XVIII.13).

There are several *theoretical* issues which require further study. The most interesting points seem to be: (i) More detailed models for the surface melting. In particular, one would like to include the decay of translational order perpendicular to the surface which has been ignored in Ref. (XVIII.28). This problem can be addressed by density functional theory [XVIII.33,XVIII.35]. (ii) The influence of long–range oscillatory interactions in metals and alloys. (iii) The influence of interfacial fluctuations which I have not addressed in this paper. For the direct interaction as given by (XVIII.6) or (XVIII.7) , interfacial fluctuations are expected to lead to a non–trivial renormalization of the critical behavior [XVIII.24,XVIII.21,XVIII.36]. It is not clear, however, if these fluctuation effects will be visible in experiments, since computer simulations of lattice models have not produced, so far, any evidence for them [XVIII.37,XVIII.38].

Needles to say that more *experimental* work is highly desirable. A very promising tool is X–ray diffraction under total external reflection [XVIII.39] which has already been applied to $Cu_3Au$ [XVIII.8]. In particular, it would be interesting to look for surface–induced disorder in magnetic materials with first–order phase transitions.

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XVIII.18 For simplicity, it is tacitly assumed here that $V(\ell)$ has a unique minimum. Beyond mean-field theory, $V_{DI}(\ell)$ must be replaced by a renormalized interaction which includes the effect of interfacial fluctuations. Quite generally, these fluctuations act to separate the (do) interface from the (vd) interface and, thus, to enhance the tendency towards complete surface-induced disorder (or surface melting).

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