Quadratic substrate energy term and harmonics in the Halperin-Nelson model

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The influence of the quadratic term in the Taylor expansion for the substrate potential, which is neglected in the Halperin-Nelson model, is investigated. The quadratic term is found to yield a harmonic series for the equilibrium displacement and a quadratic potential term in the energy of deviations from this displacement. Criteria for convergence of the series and negligibility of the quadratic potential term are derived. The criteria require the misfit vernier period and length scale of deviations not to be large compared with the Van der Merwe discommensuration width.

INTRODUCTION

The study of dislocation unbinding or Kosterlitz-Thouless transitions in two dimensions is of theoretical and practical interest. Current theories are based on the work done by Kosterlitz and Thouless[1] (KT) and Young[2] for free layers, and by Halperin and Nelson[3] (HN) who included an incommensurate modulating layer-substrate potential (MP).

HN, using a linear response approximation, discarded the term in the Taylor expansion of the MP that is quadratic in displacements from the free condition of the adlayer, and higher-order terms. In this approximation, it is possible to eliminate the explicit occurrence of the MP in the energy of the adlayer by introducing a shift in the reciprocal space representation of the displacement. This corresponds to a sinusoidal displacement, which represents an equilibrium state between the competing periodicities of the elastic force and the truncated MP. We will refer to this displacement as the regular displacement. Henceforth the deviation from the regular displacement will be termed the residual displacement. The energy of the system then has a regular elastic form in the residual displacement with an additional term dependent on the relative orientation of the adlayer and the substrate added. This energy can then be treated in the framework of KT theory.

This linear-response approximation is subject to the restrictions that the MP should not be too strong (or that the temperature be high enough) and that the adlattice be sufficiently incommensurate with the substrate.[4] In order to quantify the above restrictions, we study the effect on the HN model of the quadratic term in the Taylor expansion of the MP. The main effects of the quadratic term are found to be the introduction of higher-order harmonics of the regular displacement found by HN, similar to the charge-density-wave harmonics predicted by McMillan[5] and Nakanishi and Shiba,[6] and the persistence of a quadratic MP term dependent on the residual displacement.

Our main goals are to assess the effect of the quadratic term and to quantify the restrictions this imposes on the linear-response approximation of the HN model.

THE HN MODEL

The HN model comprises an isotropically elastic monolayer (ML) adsorbed on a rigid substrate, in which the ML-substrate interaction is represented by a modulating potential (MP).[3]

The elastic energy $\mathcal{H}_E$ of the ML is given by (summation convention is implied by repeated indices)

$$\mathcal{H}_E=\tfrac{1}{2}\int_{\mathbf{r}}\{2\mu u_{ij}(\mathbf{r})^2+\lambda[u_{kk}(\mathbf{r})]^2\}\ , \tag{1}$$

where

$$u_{ij}=\tfrac{1}{2}(\partial_i u_j+\partial_j u_i)\ . \tag{2}$$

$\mathbf{u}$ is the displacement field of ML atoms from their "free" positions, i.e., the positions they would have had, had there not been a MP, and λ and μ are the Lamé elastic constants. The energy of the ML in the MP can be expressed in terms of the reciprocal lattice vectors $\mathbf{K}$ of the substrate and the MP Fourier coefficients $g_{\mathbf{K}}$ as

$$\mathcal{H}_V=-\sum_{\mathbf{R}}\sum_{\mathbf{K}}g_{\mathbf{K}}\exp\{i\mathbf{K}\cdot[\mathbf{R}+\mathbf{u}(\mathbf{R})]\}\ , \tag{3}$$

where the $\mathbf{R}$ are the lattice vectors of the adlayer. We transform to reciprocal space by

$$\mathbf{u}(\mathbf{q})=\int_{\mathbf{R}}\mathbf{u}(\mathbf{R})e^{i\mathbf{q}\cdot\mathbf{R}}\ . \tag{4}$$

$\mathbf{u}(\mathbf{q})$ is the Fourier transform of the displacement field in the ML reciprocal lattice and $\mathbf{q}$ is a ML reciprocal lattice vector in the first Brillouin zone. The total energy of the adlayer is then given by

$$\begin{aligned}\mathcal{H}&=\mathcal{H}_E+\mathcal{H}_V\\&=\tfrac{1}{2}\int_{\mathbf{q}}u_i(\mathbf{q})D_{ij}(\mathbf{q})u_j(-\mathbf{q})\\&\quad+\sum_{\mathbf{R}}\sum_{\mathbf{K}}g_{\mathbf{K}}e^{i\mathbf{K}\cdot\mathbf{R}}\{1+i\mathbf{K}\cdot\mathbf{u}(\mathbf{R})-\tfrac{1}{2}[\mathbf{K}\cdot\mathbf{u}(\mathbf{R})]^2\cdots\}\end{aligned} \tag{5}$$

$$=\tfrac{1}{2}\int_{\mathbf{q}}\Big[\mathbf{u}(\mathbf{q})\cdot\underline{D}(\mathbf{q})\cdot\mathbf{u}(-\mathbf{q})-2\mathbf{u}^0(\mathbf{q})\cdot\sum_{\mathbf{K}}ig_{\mathbf{K}}\mathbf{K}\Delta_{\mathbf{K},-\mathbf{q}}+\mathbf{u}(\mathbf{q})\cdot\sum_{\mathbf{K}}g_{\mathbf{K}}\mathbf{K}\mathbf{K}\cdot\mathbf{u}(-\mathbf{K}-\mathbf{q})\cdots\Big]\ . \tag{6}$$

$\underline{D}(\mathbf{q})$ is the dynamical matrix of the ML. $\Delta_{\mathbf{K},\mathbf{q}}=\Sigma_{\mathbf{R}}e^{i\mathbf{K}\cdot\mathbf{R}}$ is the Dirac δ function if $\mathbf{K}=\mathbf{q}+\mathbf{G}$ for some ML reciprocal lattice vector $\mathbf{G}$; otherwise it is zero. Note that the quadratic term, dropped by HN, in the expansion of the MP is retained.

To eliminate the terms linear in $\mathbf{u}$, we write

$$\mathbf{u}(\mathbf{q})=\mathbf{u}^0(\mathbf{q})+\mathbf{v}^0(\mathbf{q}) , \tag{7}$$

where $\mathbf{u}^0$ (and later $\mathbf{u}^i$) is the residual displacement, while $\mathbf{v}^0$ (and $\mathbf{v}^i$) is the regular displacement. By introducing the shift $\mathbf{v}^0$, we hope to eliminate the influence of the MP from the energy, so that we can apply the theory of Kosterlitz and Thouless for dislocation unbinding. The displacement of the dislocation is thus included in $\mathbf{u}^i$, while the $\mathbf{v}^i$ should be a "background" term incorporating the effects of the MP. This worked well when only the linear term in the expansion of the MP was retained. What is of interest now is whether the retention of the quadratic term will still allow this decoupling of MP and dislocations. Introducing the shorthand notation

$$\mathbf{C}^0_{\mathbf{K}}(-\mathbf{q})=\sum_{\mathbf{K}} i g_{\mathbf{K}}\mathbf{K}\Delta_{\mathbf{K},-\mathbf{q}} , \tag{8}$$

$$\underline{S}_{\mathbf{K}}=\sum_{\mathbf{K}} g_{\mathbf{K}}\mathbf{K}\mathbf{K} , \tag{9}$$

and substituting in Eq. (6), we obtain after simplification

$$\mathcal{H}=\tfrac{1}{2}\int_{\mathbf{q}}[\mathbf{u}^0(\mathbf{q})\cdot\underline{D}(\mathbf{q})\cdot\mathbf{u}^0(-\mathbf{q})-2\mathbf{u}^0(\mathbf{q})\cdot\mathbf{C}^0_{\mathbf{K}}(-\mathbf{q})+\mathbf{u}^0(\mathbf{q})\cdot\underline{S}_{\mathbf{K}}\cdot\mathbf{u}^0(-\mathbf{K}-\mathbf{q})+2\mathbf{u}^0(\mathbf{q})\cdot\underline{D}(\mathbf{q})\cdot\mathbf{v}^0(-\mathbf{q})$$
$$+2\mathbf{u}^0(\mathbf{q})\cdot\underline{S}_{\mathbf{K}}\cdot\mathbf{v}^0(-\mathbf{K}-\mathbf{q})+\mathbf{v}^0(\mathbf{q})\cdot\underline{D}(\mathbf{q})\cdot\mathbf{v}^0(-\mathbf{q})-2\mathbf{v}^0(\mathbf{q})\cdot\mathbf{C}^0_{\mathbf{K}}(-\mathbf{q})+\mathbf{v}^0(\mathbf{q})\cdot\underline{S}_{\mathbf{K}}\cdot\mathbf{v}^0(-\mathbf{K}-\mathbf{q})] . \tag{10}$$

We can eliminate the terms that are linear in $\mathbf{u}^0$ by defining

$$\mathbf{v}^0(\mathbf{q})=\underline{D}^{-1}(\mathbf{q})\cdot\mathbf{C}^0_{\mathbf{K}}(\mathbf{q})=i\underline{D}^{-1}(\mathbf{q})\cdot\sum_{\mathbf{K}}\mathbf{K}g_{\mathbf{K}}\Delta_{\mathbf{K},\mathbf{q}} . \tag{11}$$

The energy of the system accordingly becomes

$$\mathcal{H}=\tfrac{1}{2}\int_{\mathbf{q}}[\mathbf{u}^0(\mathbf{q})\cdot\underline{D}(\mathbf{q})\cdot\mathbf{u}^0(-\mathbf{q})+\mathbf{u}^0(\mathbf{q})\cdot\underline{S}_{\mathbf{K}}\cdot\mathbf{u}^0(-\mathbf{K}-\mathbf{q})+2\mathbf{u}^0(\mathbf{q})\cdot\underline{S}_{\mathbf{K}}\cdot\mathbf{v}^0(-\mathbf{K}-\mathbf{q})$$
$$-\mathbf{v}^0(\mathbf{q})\cdot\underline{D}(\mathbf{q})\cdot\mathbf{v}^0(-\mathbf{q})+\mathbf{v}^0(\mathbf{q})\cdot\underline{S}_{\mathbf{K}}\cdot\mathbf{v}^0(\mathbf{K}-\mathbf{q})] . \tag{12}$$

We now define

$$\mathbf{C}^1_{\mathbf{K}}(-\mathbf{q})=-\underline{S}_{\mathbf{K}}\cdot\mathbf{v}^0(-\mathbf{K}-\mathbf{q}) . \tag{13}$$

Because $\mathbf{v}^0$ is defined in Eq. (11) in terms of the MP and adlayer elasticity, the last two terms in Eq. (12) are effectively constants. Equation (12) therefore is of exactly the same form as (10). We can thus recursively eliminate terms that are linear in $\mathbf{u}^{n-1}$ by defining

$$\mathbf{u}^{n-1}(\mathbf{q})=\mathbf{u}^n(\mathbf{q})+\mathbf{v}^n(\mathbf{q}) , \tag{14a}$$

$$\mathbf{v}^n(\mathbf{q})=\underline{D}^{-1}(\mathbf{q})\cdot\mathbf{C}^n_{\mathbf{K}}(\mathbf{q}) , \tag{14b}$$

and

$$\mathbf{C}^n_{\mathbf{K}}(-\mathbf{q})=-\underline{S}_{\mathbf{K}}\mathbf{v}^{n-1}(-\mathbf{K}-\mathbf{q}) . \tag{14c}$$

Repeating the recursion until convergence, the energy can be shown to be

$$\mathcal{H}=\tfrac{1}{2}\int_{\mathbf{q}}\Bigg\{\mathbf{u}^\infty(\mathbf{q})\cdot\underline{D}(\mathbf{q})\cdot\mathbf{u}^\infty(-\mathbf{q})+\mathbf{u}^\infty(\mathbf{q})\cdot\underline{S}_{\mathbf{K}}\cdot\mathbf{u}^\infty(-\mathbf{K}-\mathbf{q})$$
$$+\sum_{n=0}^{\infty}[-\mathbf{v}^n(\mathbf{q})\cdot\underline{D}(\mathbf{q})\cdot\mathbf{v}^n(-\mathbf{q})+\mathbf{v}^n(\mathbf{q})\cdot\underline{S}_{\mathbf{K}}\cdot\mathbf{v}^n(-\mathbf{K}-\mathbf{q})]\Bigg\} . \tag{15}$$

In this limit, the constant terms decouple from the residual displacement field $\mathbf{u}^\infty$. Only the elastic and quadratic MP components of the energy remain.

To interpret Eq. (15), we write $\mathbf{v}^n$ explicitly as

$$\mathbf{v}^{n+1}(\mathbf{q})=(-1)^n\prod_{j=0}^{n}\left[\underline{D}^{-1}\left[\sum_{l=1}^{j}\mathbf{K}^l+\mathbf{q}\right]\cdot\underline{S}_{\mathbf{K}^{l'}}\right]i\underline{D}^{-1}\left[\sum_{l=0}^{n}\mathbf{K}^l+\mathbf{q}\right]\sum_{\mathbf{K}}g_{\mathbf{K}}\mathbf{K}\Delta_{\sum_{l=1}^{m}\mathbf{K}^l+\mathbf{K},\mathbf{q}} . \tag{16}$$

Because of the Δ, this equation shows the $\mathbf{v}^n(\mathbf{r})$ to be Fourier sums over such reciprocal lattice vectors $\mathbf{p}$ of the first Brillouin zone, as correspond to a difference between a reciprocal lattice vector $\mathbf{G}$ of the ML and an n-fold composition of the substrate reciprocal lattice vectors $\mathbf{K}$ contained in the expansion of the potential. These $\mathbf{p}=\sum_l \mathbf{K}-\mathbf{G}$ are misfit vectors of order n. This selection criterion on displacement modulation frequencies corresponds to the criterion for epitaxial configuration selection and discommensuration (DC) spacing derived by Braun and Van der Merwe.[7] The $\mathbf{v}^n(\mathbf{p})$ are just the Fourier components of the displacement field of a regular DC lattice taking up all the misfits, similar to the expansions for charge-density waves developed by McMillan[4] and Nakanishi and Shiba.[5] The $\mathbf{u}^n(\mathbf{q})$ are the Fourier components of the deviation of the displacement field from this regular DC lattice.

Substituting (16) into (15) and writing the dynamical matrix in terms of polarization vectors $\epsilon_s(\mathbf{p})$, the mass of the adatom m and eigenfrequencies $\omega(\mathbf{p})$, we obtain

$$\mathcal{H}=\frac{1}{2}\int_{\mathbf{q}}[\mathbf{u}^{\infty}(\mathbf{q})\cdot\underline{D}(\mathbf{q})\cdot\mathbf{u}^{\infty}(-\mathbf{q})+\mathbf{u}^{\infty}(\mathbf{q})\cdot\underline{S}_{\mathbf{K}}\cdot\mathbf{u}^{\infty}(-\mathbf{K}-\mathbf{q})]$$
$$+\sum_{n=2}^{\infty}\sum_{\{\mathbf{K}_i^n\}^*}(-1)^n g_{\mathbf{K}_i^n}\prod_{l=2}^{n}\left|g_{\mathbf{K}_i^n}\sum_{s=1}^{2}\frac{\left[\mathbf{K}_{l-1}^n\cdot\epsilon_s\left[\sum_{i=1}^{l}\mathbf{K}_i^n\right]\right]\left[\mathbf{K}_l^n\cdot\epsilon_s\left[\sum_{i=1}^{l}\mathbf{K}_i^n\right]\right]}{m\omega^2\left[\sum_{i=1}^{l}\mathbf{K}_i^n\right]}\right| . \tag{17}$$

$\{\mathbf{K}_i^n\}^*$ denotes the n-fold stars of substrate reciprocal vectors, i.e., all those sets of n vectors summing to 0. As in the original HN model, the sum in (17) depends only on the relative orientation of substrate and adlayer. If this series converges, the greater number of terms than in the original HN model does not substantially alter their conclusions. However, the second term in the integral, representing the effect of the MP on deviations from the regular displacement modulations $\mathbf{v}^i$, does not appear in the HN model.

CRITERIA

The effect of the quadratic term in the expansion of the modulation potential is shown to be twofold: (a) The regular displacement modulation is a Fourier series including higher-order harmonics. (b) There remains in the expression for the energy a MP term dependent on the residual displacement. These effects enable us to determine criteria for the applicability of HN theory. If both effects are small, i.e., if the series in (15) converges and if the remaining MP term is much smaller than the elastic contribution to the energy, HN will hold true.

To derive a good approximate criterion for the convergence of the series in (15), we note that

$$\underline{D}(\mathbf{q})\cong\mu q^2\mathbb{1}+(\lambda+\mu)\mathbf{q}\mathbf{q} \tag{18}$$

so that

$$\|\mathbf{v}^n(\mathbf{p})\|/\|\mathbf{v}^{n-1}(\mathbf{p})\|\approx l^{-2}p^{-2} , \tag{19}$$

where

$$l^{-2}=\left|\left|\sum_{\mathbf{K}}g_{\mathbf{K}}\mathbf{K}\mathbf{K}/(2\mu+\lambda)\right|\right| . \tag{20}$$

This series converges provided

$$p^{-1}\le l . \tag{21}$$

In Eq. (16) for $\mathbf{v}^n$, those $\mathbf{p}$'s for which $\Delta\neq 0$ are reciprocal misfit vectors and determine the spacing between "discommensurations." l corresponds to the Van der Merwe l-parameter,[8,9] giving the width of a DC. Condition (21) thus requires the DC spacing to be less than, or of the order of, the DC width or equivalently requires the misfit to be large and the MP weak compared with the elastic constants. This implies that, near a commensurate-incommensurate transition, many terms of the expansion will have to be included if convergence can be attained at all.[4,5]

In addition, (15) shows that the potential terms, except the linear term, carry across to the modified $\mathbf{u}(\mathbf{q})$. The HN theory, which disregards these terms, is valid if they are negligible. Estimating their size relative to the elastic terms we obtain

$$\mathbf{u}^{\infty}(\mathbf{q})\cdot\underline{S}_{\mathbf{K}}(\mathbf{q})\cdot\mathbf{u}^{\infty}(-\mathbf{K}-\mathbf{q})/[\mathbf{u}^{\infty}(\mathbf{q})\cdot\underline{D}(\mathbf{q})\cdot\mathbf{u}^{\infty}(\mathbf{q})]\approx q^{-2}l^{-2} . \tag{22}$$

In this equation, $\mathbf{q}$ is the wave vector of the perturbations being considered. According to Eq. (22), for the MP influence on the modified $\mathbf{u}$ to be small relative to the elastic energy of this displacement field requires the characteristic distance q^{-1} of these perturbations to be small compared with the DC width l. This, the exact analogy of (21), shows the MP to be especially important when considering long-range interactions.

CONCLUSIONS

Investigating the influence of the MP quadratic term in the HN model, we found the MP induced regular displacement consists of a Fourier series. The linear approximation used by Halperin and Nelson gives only the first term of this series. The energy includes a term quadratic in the residual displacement. Criteria were found for the convergence of the series and the smallness of the quadratic potential term in the residual displacement. These criteria limit the range of applicability of the linear-response approximation used in HN theory. The criteria are as follows.

(a) The misfit vernier periods must be of the order of the discommensuration width (as given by the Van der Merwe l parameter). This requires discommensurations to be closely spaced and therefore requires commensurate-incommensurate transition to have advanced far.

(b) The inverse of the wave vector of disturbances considered must be of the order of the discommensuration width.

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