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Phase transitions in solids of diperiodic symmetry

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All isotropy subgroups (and thus all quasicontinuous symmetry changes) corresponding to **k** points of symmetry have been obtained for the 80 diperiodic space groups. The detailed information for such phase transitions is given here for the diperiodic space group $P\frac{4}{m}\frac{2}{m}\frac{2}{m}$. Only two distinct images (sets of representation matrices) occur for this example yielding Landau-Ginzburg-Wilson (LGW) Hamiltonians corresponding to the Ising and XY models, respectively. Minimization of the LGW Hamiltonians yields those transitions which are continuous in the mean-field description.

I. INTRODUCTION

A widely used initial approach to the description of phase transitions is the phenomenological Landau theory.[1] The theory has been well used in three-dimensional systems and extensions[2] of the original formulation have made it possible to distinguish a set of general direct group-theoretical necessary conditions as well as the usual condition which minimizes the free energy. The group-theoretical conditions lead to the selection of isotropy subgroups by means of the subduction and chain criteria[3] from which the Landau and Lifshitz conditions further select. Recently a systematic method for obtaining isotropy groups of a space group G_0 was discussed[4] and then used to obtain all isotropy groups of the 230 three-dimensional space groups.[5]

The listing of isotropy subgroups provides a complete list of all possible broken-symmetry phases that can occur as a result of minimization of a general invariant thermodynamic free energy. We need not restrict attention to lower-order expansions in terms of the vector order parameter (as in the original Landau description) but may also consider first-order transitions corresponding to a more general free-energy and thus "quasicontinuous" first-order transitions. All such resulting lower-symmetry phases are the isotropy subgroups corresponding to the irreducible representation of the order parameter in which the free energy is expressed. Thus, first-order or multicritical transitions to lower phases are given by the isotropy subgroup listing.

Order-disorder transitions in physisorbed and chemisorbed surfaces[6] as well as structural surface transitions[7] in clean metals have caused a great deal of attention to be focused on two-dimensional transitions in the last several years. Recent detailed experiments [low-energy electron diffraction (LEED), Auger-electron spectroscopy, etc.] have justified a significant amount of previous theoretical modeling for two-dimensional systems[6] and have promised significant contributions for the future. With the need apparent, several authors classified transitions in two-dimensional systems.[8] Our algorithm for obtaining isotropy groups was applied to the 17 two-dimensional space groups and the complete listing of quasicontinuous phase transitions published.[9]

Of the transitions associated with surfaces many are reversible with temperature[10] and correspond to continuous transitions. The symmetry of a surface system is that of the surface layer *and* the substrate (or semi-infinite bulk). The symmetry group would then be one of the 17 two-dimensional space groups. Our previous listing[9] classifies the possibilities of such transitions. However, for a layer weakly coupled to the bulk, the symmetry can include more symmetry than allowed by the 17 two-dimensional space groups. It has been shown[11] in the layer structure As_2S_3 that pressure affects the interlayer coupling causing a change from more-free layers to more interbonded layers. The more-free-layer spectra were seen to correspond to diperiodic symmetry. This change from diperiodic space-group symmetry to three-dimensional symmetry is evidence of the need to use diperiodic space groups for the correct description of the symmetry in nearly free layers. The diperiodic symmetry is of course approximate for a bonded layer but becomes exact as the layer becomes isolated. The resulting 80 diperiodic space groups have been listed[12] and necessarily contain the two-dimensional space groups as a subset.

Changes in translational symmetry for the diperiodics have been reported[13] as well as a listing of transitions which conserve the number of atoms per unit cell.[14] However, general symmetry transitions which allow translational and point changes have not been published. We report here that *all space group changes for quasicontinuous transitions* (corresponding to **k** points of symmetry) *in the diperiodic space groups have now been obtained.* Below we outline the method for obtaining isotropy subgroups of space groups and then list as an example the symmetry changes corresponding to the diperiodic space group $P\frac{4}{m}\frac{2}{m}\frac{2}{m}$ (which is not one of the 17 two-dimensional space groups). The results for all the diperiodics is a lengthy table and will be published elsewhere. We construct Landau-Ginzburg-Wilson (LGW) Hamiltonians for all representations which satisfy the Lifshitz condition for this space group and as a result, the universality classes are indicated. Minimization of these fourth-order LGW Hamiltonians then selects those transitions which could be second-order within Landau theory.

Renormalization-group methods have the more definitive word as to whether the transitions remain continuous when fluctuations are included.

II. ISOTROPY SUBGROUPS, MINIMIZATION

For the space group $G_0=P\frac{4}{m}\frac{2}{m}\frac{2}{m}$, transitions will be determined by the multicomponent order parameter (basis) of each irreducible representation of G_0. Considering one representation of G_0, say $D^{(*\mathbf{k},m)}$, an isotropy subgroup G will be the largest subgroup of G_0 leaving a subspace (call it Fix G) of the representation space fixed, i.e., $g\boldsymbol{\psi}=\boldsymbol{\psi}$ for g in G, and $\boldsymbol{\psi}$ in the subspace Fix G. The dimensionality of the subspace Fix G and its corresponding isotropy subgroup for all such subspaces of $D^{(*\mathbf{k},m)}$ can be obtained group theoretically from the characters of the representation through the subduction and chain criteria.[3] The selection of isotropy groups becomes systematic[4] as translation subgroups T_G $(T_G\subset T_0)$ and point groups P_G $(P_G\subset P_0)$ are selected such that they satisfy the subduction condition, the selection forms an extension of T_G by P_G, and the chain criterion is satisfied. A nontrivial part of the isotropy subgroup listing is to identify the subgroup in standard form[15] and to indicate relative origin and orientation selection. This last information is of crucial importance in detailed microscopic considerations associated with experiments (x-ray, NMR, LEED, etc.).

In Table I we list for the diperiodic group $P\frac{4}{m}\frac{2}{m}\frac{2}{m}$ the irreducible representation, Landau frequency (number of third-order invariants), Lifshitz frequency (number of two-dimensional vector representations in the antisymmetric square), subduction frequency, isotropy subgroup, new lattice basis, and new origin selection. The lattice basis and origin selection are given in terms of the original primitive vectors. This information was determined as outlined above. We follow the notation of Ref. 16 in labeling representations and use the spacegroup designations of Ref. 12.

Methods which take into account fluctuations have shown that the Landau condition (Landau frequency equal to zero) is not a necessary condition in two-dimensional systems[14,17] but that the Liftshitz condition (Lifshitz frequency equal to zero) is necessary when considering commensurate transitions. Restricting our attention to those representations which do satisfy the Lifshitz condition all resulting representations are of one or two dimensions for our example. The dimensionality of a particular representation can be obtained from the highest subduction frequency appearing in Table I for that representation.

All one-dimensional representations (other than the identity representation Γ_1^+ which causes no symmetry breaking) yield the LGW Hamiltonian

$$F=\sum_{\alpha=1}^{3} u_\alpha I_\alpha , \qquad (1)$$

with $I_1=\psi^2$, $I_2=I_1^2$, and $I_3=I_1^3$. We have restricted the expansion to sixth order since we are primarily interested in continuous transitions from the high-symmetry phase within this mean-field description. Usual minimization procedures[1] yield a continuous transition from $P\frac{4}{m}\frac{2}{m}\frac{2}{m}$ at $u_1=0$ and for $u_2>0$. Only one lower-symmetry phase is possible corresponding to those group elements which leave ψ fixed. The equilibrium Hamiltonian goes as $F=-u_1^2/4u_2$ as $u_1\to 0$ with $\psi^2=-u_1/2u_2$. Thus this continuous transition could occur along a line in pressure, temperature (P,T) variables. The LGW Hamiltonian is in the Ising universality class and has been well studied by renormalization-group methods yielding exponents $\beta=\frac{1}{8}$, $\gamma=\frac{7}{4}$, $\nu=1$, and $\alpha=0$.

All two-dimensional representations of $P\frac{4}{m}\frac{2}{m}\frac{2}{m}$ have the same image, i.e., the same set of distinct representation matrices. The set of representation matrices (eight matrices for this example) are isomorphic to the point group in two dimensions C_{4v}. The resulting LGW Hamiltonian is thus the same for each of these representations as well as the minimization process and universality classification. We need only consider isotropy subgroups of the image C_{4v} in determining isotropy space subgroups. The corresponding space groups will be obtained from the inverse images of the matrix isotropy subgroups. For example, the two-dimensional representation corresponding to Γ_5^+ has three matrix isotropy subgroups of C_{4v}, namely the subgroups $m(y)$, $m(d)$, and 1. (Here the letters y and d indicate the orientation of the mirror planes in representation space.) The isotropy subgroup $m(y)$ leaves the one-dimensional subspace $(\psi_1\neq 0,\psi_2=0)$ fixed and its inverse image yields phase $P11\frac{2}{m}$. Isotropy subgroup $m(d)$ leaves fixed the one-dimensional subspace $(\psi_1=\psi_2\neq 0)$ and yields phase $C11\frac{2}{m}$. The third isotropy subgroup 1, corresponds to the two-dimensional subspace (generic points) $(\psi_1\neq\psi_2$, with $\psi_1\neq 0$, $\psi_2\neq 0)$ and yields phase $P\bar{1}$. Corresponding to each of the two-dimensional representations of $P\frac{4}{m}\frac{2}{m}\frac{2}{m}$ we will obtain three isotropy subgroups G_1, G_2, and G_3 from inverse images of $m(y)$, $m(d)$, and 1, respectively. The inverse images will of course depend upon the specific representation considered, as can be seen from Table I.

The LGW Hamiltonian for the two-dimensional representations of $P\frac{4}{m}\frac{2}{m}\frac{2}{m}$ is of the form

$$F=\sum_{\alpha=1}^{3} u_\alpha I_\alpha+\sum_{\beta=1}^{2} v_\alpha J_\alpha , \qquad (2)$$

with $I_1=\psi_1^2+\psi_2^2$, $I_2=I_1^2$, $I_3=I_1^3$, $J_1=\psi_1^4+\psi_2^4$, and $J_2=I_1J_1$. This Hamiltonian is the XY model with cubic anisotropy.

Using a method introduced in the analysis of the Higgs potential,[18] the minimization of this Hamiltonian can be easily obtained. Defining $\psi_1^2+\psi_2^2=\psi^2$ and $\lambda=(\psi_1^4+\psi_2^4)/\psi^4$, we obtain

$$F=u_1\psi^2+u_2'\psi^4+u_3'\psi^6 , \qquad (3)$$

with $u_2'=u_2+\lambda v_1$ and $u_3'=u_3+\lambda v_2$. The above Hamiltonian is formally the same as for the above Ising model except the coefficients u_2' and u_3' are now functions of λ. Thus along a direction specified by λ we can obtain a directional minimum[18] by setting $\partial F/\partial\psi=0$ and requiring $\partial^2F/\partial^2\psi>0$. In order to find the *absolute* minimum of F we look for the lowest minimum among the directional minima (which are a function of λ), i.e., we also consider

TABLE I. Isotropy subgroups G of the diperiodic space group $P\frac{4}{m}\frac{2}{m}\frac{2}{m}$. We give the Landau frequency $[D^3]$, the Lifshitz frequency $\{D^2\}_v$, the subduction frequency $i(G)$, the primitive basis vectors, $\mathbf{t}_1$ and $\mathbf{t}_2$, in terms of the primitive basis vectors of G_0, and the change τ in space-group origin (from G_0 to G) in terms of the primitive basis vectors of G_0.

irrep	$[D^3]$	$\{D^2\}_v$	$i(G)$	G	$\mathbf{t}_1$	$\mathbf{t}_2$	τ
Γ_1^+	1	0	1	$P\frac{4}{m}\frac{2}{m}\frac{2}{m}$	1,0	0,1	0,0
Γ_2^+	0	0	1	$P\frac{2}{m}\frac{2}{m}\frac{2}{m}$	1,0	0,1	0,0
Γ_3^+	0	0	1	$P\frac{4}{m}$	1,0	0,1	0,0
Γ_4^+	0	0	1	$C\frac{2}{m}\frac{2}{m}\frac{2}{m}$	0,1	-1,0	0,0
Γ_5^+	0	0	1	$C11\frac{2}{m}$	1,0	0,1	0,0
			1	$P11\frac{2}{m}$	0,1	-1,0	0,0
			2	$P\bar{1}$	1,0	0,1	0,0
Γ_1^-	0	0	1	$P422$	1,0	0,1	0,0
Γ_2^-	0	0	1	$P\bar{4}2m$	1,0	0,1	0,0
Γ_3^-	0	0	1	$P4mm$	1,0	0,1	0,0
Γ_4^-	0	0	1	$P\bar{4}m2$	1,0	0,1	0,0
Γ_5^-	0	0	1	$Cmm2$	0,1	-1,0	0,0
			1	$Pmm2$	1,0	0,1	0,0
			2	$Pm11$	1,0	0,1	0,0
M_1^+	0	0	1	$P\frac{4}{m}\frac{2}{m}\frac{2}{m}$	1,1	-1,1	0,0
M_2^+	0	0	1	$P\frac{4}{m}\frac{2_1}{b}\frac{2}{m}$	1,1	-1,1	$\frac{1}{2}$,$\frac{1}{2}$
M_3^+	0	0	1	$P\frac{4}{m}\frac{2_1}{b}\frac{2}{m}$	1,1	-1,1	0,0
M_4^+	0	0	1	$P\frac{4}{m}\frac{2}{m}\frac{2}{m}$	1,1	-1,1	$\frac{1}{2}$,$\frac{1}{2}$
M_5^+	0	0	1	$C\frac{2}{a}\frac{2}{m}\frac{2}{m}$	1,1	-1,1	$\frac{1}{2}$,$\frac{1}{2}$
			1	$P\frac{2}{n}\frac{2}{m}\frac{2_1}{a}$	1,-1	1,1	0,0
			2	$P\frac{2}{b}11$	1,-1	0,2	0,0
M_1^-	0	0	1	$P\frac{4}{n}\frac{2}{b}\frac{2}{m}$	1,1	-1,1	0,0
M_2^-	0	0	1	$P\frac{4}{n}\frac{2_1}{m}\frac{2}{m}$	1,1	-1,1	0,0
M_3^-	0	0	1	$P\frac{4}{n}\frac{2_1}{m}\frac{2}{m}$	1,1	-1,1	$\frac{1}{2}$,$\frac{1}{2}$
M_4^-	0	0	1	$P\frac{4}{n}\frac{2}{b}\frac{2}{m}$	1,1	-1,1	$\frac{1}{2}$,$\frac{1}{2}$
M_5^-	0	0	1	$C\frac{2}{m}\frac{2}{m}\frac{2}{m}$	1,1	-1,1	$\frac{1}{2}$,0
			1	$P\frac{2}{m}\frac{2_1}{m}\frac{2}{a}$	1,-1	1,1	$\frac{1}{2}$,0
			2	$P\frac{2}{m}11$	1,1	-1,1	0,$\frac{1}{2}$
X_1^+	0	0	1	$P\frac{2}{m}\frac{2}{m}\frac{2}{m}$	2,0	0,1	0,0
			1	$P\frac{4}{m}\frac{2}{m}\frac{2}{m}$	2,0	0,2	0,0
			2	$P\frac{2}{m}\frac{2}{m}\frac{2}{m}$	2,0	0,2	0,0
X_2^+	0	0	1	$P\frac{2}{m}\frac{2_1}{m}\frac{2}{a}$	2,0	0,1	0,0
			1	$P\frac{4}{m}\frac{2_1}{b}\frac{2}{m}$	2,0	0,2	0,0
			2	$P\frac{2}{m}\frac{2_1}{b}\frac{2_1}{a}$	2,0	0,2	0,0
X_3^+	0	0	1	$P\frac{2}{a}\frac{2_1}{m}\frac{2}{m}$	2,0	0,1	0,0
			1	$P\frac{4}{n}\frac{2_1}{m}\frac{2}{m}$	2,0	0,2	$\frac{1}{2}$,$\frac{1}{2}$
			2	$P\frac{2}{n}\frac{2_1}{m}\frac{2_1}{m}$	2,0	0,2	$\frac{1}{2}$,$\frac{1}{2}$
X_4^+	0	0	1	$P\frac{2}{a}\frac{2}{m}\frac{2}{a}$	2,0	0,1	0,0
			1	$P\frac{4}{n}\frac{2}{b}\frac{2}{m}$	2,0	0,2	$\frac{1}{2}$,$\frac{1}{2}$
			2	$P\frac{2}{n}\frac{2}{b}\frac{2}{a}$	2,0	0,2	$\frac{1}{2}$,$\frac{1}{2}$

TABLE I. (*Continued*).

irrep	$[D^3]$	$\{D^2\}_v$	$i(G)$	G	$\mathbf{t}_1$	$\mathbf{t}_2$	$\boldsymbol{\tau}$
X_1^-	0	0	1	$P\frac{2}{a}\frac{2}{m}\frac{2}{a}$	2,0	0,1	$\frac{1}{2}$,0
			1	$P\frac{4}{n}\frac{2}{b}\frac{2}{m}$	2,0	0,2	0,0
			2	$P\frac{2}{n}\frac{2}{b}\frac{2}{a}$	2,0	0,2	0,0
X_2^-	0	0	1	$P\frac{2}{a}\frac{2_1}{m}\frac{2}{m}$	2,0	0,1	$\frac{1}{2}$,0
			1	$P\frac{4}{n}\frac{2_1}{m}\frac{2}{m}$	2,0	0,2	0,0
			2	$P\frac{2}{n}\frac{2_1}{m}\frac{2_1}{m}$	2,0	0,2	0,0
X_3^-	0	0	1	$P\frac{2}{m}\frac{2_1}{m}\frac{2}{a}$	2,0	0,1	$\frac{1}{2}$,0
			1	$P\frac{4}{m}\frac{2_1}{b}\frac{2}{m}$	2,0	0,2	$\frac{1}{2}$,$\frac{1}{2}$
			2	$P\frac{2}{m}\frac{2_1}{b}\frac{2_1}{a}$	2,0	0,2	$\frac{1}{2}$,$\frac{1}{2}$
X_4^-	0	0	1	$P\frac{2}{m}\frac{2}{m}\frac{2}{m}$	2,0	0,1	$\frac{1}{2}$,0
			1	$P\frac{4}{m}\frac{2}{m}\frac{2}{m}$	2,0	0,2	$\frac{1}{2}$,$\frac{1}{2}$
			2	$P\frac{2}{m}\frac{2}{m}\frac{2}{m}$	2,0	0,2	$\frac{1}{2}$,$\frac{1}{2}$

$$\frac{\partial F}{\partial \lambda}=v_1\psi^4+v_2\psi^6 . \qquad (4)$$

We see from Eq. (4) that minima of F will correspond to boundary values of λ. The range of values of λ is called the orbit space. For C_{4v}, the orbit space consists of the interval $\frac{1}{2}\le\lambda\le 1$, with $\lambda=1$ corresponding to phase $m(y)$, $\lambda=\frac{1}{2}$ to $m(d)$, and $\frac{1}{2}<\lambda<1$ to the generic phase 1. For continuous transitions as u_1 goes through zero, the lower-symmetry phase will be determined by the minimum of $F\to -u_1^2/4u_2'$ or equivalently by the sign of v_1 in $u_2'=u_2+\lambda v_1$. Thus, for $v_1>0$, phase $m(d)$ (G_2) is stable while for $v_1<0$, phase $m(y)$ (G_1) is stable. Notice that phase 1 (G_3) cannot be selected by a continuous transition from the high-symmetry phase. Considering F to eighth order gives the additional terms u_4I_4, $w_1I_1^2J_1$, and $w_2J_1^2$. For the extrema of F, we impose $\partial F/\partial\psi=0$ and $\partial F/\partial\lambda=0$. This second condition yields

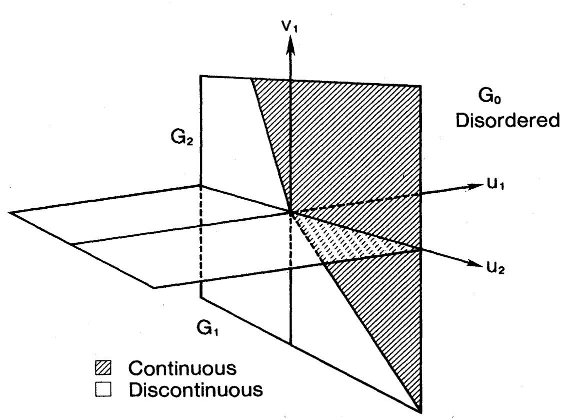

FIG. 1. Phase diagram associated with the XY model Hamiltonian. Only the region near the disordered phase is shown. For $v_1>0$, a continuous transition from G_0 to G_2 can occur if $u_2+v_1/2>0$. For $v_1<0$, a continuous transition to G_1 can occur if $u_2+v_1>0$. The transition between G_1 and G_2 is discontinuous.

$$\lambda=-(1/2w_2)(v_1/\psi^4+v_2/\psi^2+w_1)$$

and we again see that ψ cannot go to zero for $\frac{1}{2}<\lambda<1$ unless $v_1=v_2=0$, which imposes three equations to be satisfied by P and T, namely $u_1(P,T)=v_1(P,T)=v_2(P,T)=0$. Thus no continuous transition to 1 is allowed (even at a point in P,T). The above results for the mean-field XY model is compatible with the results of Refs. 19 and 20. We thus have the transition surface as indicated in Fig. 1 near the high-symmetry phase.

Renormalization-group methods near two dimensions determine if transitions corresponding to phases $m(y)$ and $m(d)$ remain continuous as fluctuations are included. The critical behavior of the XY model has been extensively studied and in particular it is known[21] that in two dimensions the critical behavior depends on the interaction (nonuniversality is evidenced). The exponents are continuous functions of the anisotropy coefficient v_1. Thus, no specific predictions will be made in this case in regards to exponents, etc.

In conclusion, we have obtained all symmetry changes corresponding to **k** points of symmetry associated with quasicontinuous transitions for the diperiodic space groups. The detailed information of such phase transitions was given here only for the diperiodic space group $P\frac{4}{m}\frac{2}{m}\frac{2}{m}$. For representations of this group satisfying the Lifshitz condition we have indicated the resulting LGW Hamiltonians and have obtained for this example two Hamiltonian forms, namely the Ising and XY models. Within the mean-field description, the phase diagram for the XY model was indicated for continuous transitions from the high-symmetry phase. Similar results for all 80 diperiodic space groups will be published elsewhere.

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