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# Symmetry change in continuous phase transitions in two-dimensional systems

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A complete listing is given of all two-dimensional lower-symmetry space groups which can arise by continuous phase transition from a parent two-dimensional space group, by breaking both rotational and translational symmetries. The group-theoretical subduction criteria are used in conjunction with a "zellengleich-klassengleich" method for listing subgroup chains. Order parameters which are considered belong to a single irreducible representation. For all cases direct group-theory results are in agreement with those obtained by minimization of a Landau free-energy polynomial, at considerable saving in effort. This listing extends the previous work of Ipatova *et al.* [Surf. Sci. 110, 543 (1981)], who only considered translational breaking. An experiment on phase transitions on the W{100} surface agrees with our results.

## I. INTRODUCTION

There recently has been increasing theoretical and experimental attention given to phase transitions in two-dimensional systems. On the theoretical side, some reasons for this include the important effect of fluctuations and the possibility of investigating exactly solvable quantum-mechanical and field-theoretical two-dimensional models. Experimentally, two-dimensional systems are important owing to studies of the structure of overlayers, adatom or admolecule arrays, surface reconstruction, and reactions on surfaces. An extensive review was recently given by Barber[1] which contains many citations to both experimental and theoretical work. Rottman[2] has given a complete listing of all universality classes of transitions in two dimensions by enumerating for each of the 17 two-dimensional (plane) crystallographic space groups "Lifshitz-active physically irreducible representations" and the associated Landau-Ginzburg-Wilson (LGW) Hamiltonians. This has extended previous work and provided a basis for study of critical behavior in such systems by renormalization-scaling methods.

One aspect of phase transitions in two-dimensional systems which has received comparatively little attention concerns the symmetry change from an initial two-dimensional space group to a final two-dimensional space group. The present paper is concerned with such symmetry change.

## II. LANDAU THEORY AND SUBDUCTION METHOD

The specific question to be studied in this paper is the following: Given a system with a symmetry group $G_0$, to which subgroups $\bar{G},\bar{G}',\ldots,$ of $G_0$ is a continuous transition permitted using the Landau theory? Here all groups $G_0,\bar{G},\bar{G}',\ldots,$ are among the seventeen two-dimensional space groups.

Classical Landau[3] theory gives a procedure for deciding the answers to this question. As reviewed by Lyubarskii[4] and by one of us,[5] Landau's theory of symmetry breaking entails these steps: (1) Determine the "active" irreducible representations $D^j_{G_0}$ of $G_0$; (2) construct the $D^j_{G_0}$-invariant Landau free energy $\Phi(\{C^j_\alpha\})$, where $\{C^j_\alpha\}$ are a set of order parameters which can be taken as bases of $D^j_{G_0}$; (3) extremize $\Phi(\{C^j_\alpha\})$ by finding the set of solutions $\{\bar{C}^j_\alpha\}$ of the equations $(\partial\Phi/\partial C^j_\beta)\,|_{(\bar{C})}=0$, where $\beta=1,\ldots,l_j$

TABLE I. Notation for the irreducible representations $D^j_{G_0}$ of parent group $G_0$. This notation applies for Tables II–XVII.

| $D^j_{G_0}$ | Condition |
|---|---|
| $D$ | Both Landau and Lifshitz active |
| $\bar{D}$ | Not Landau active, but Lifshitz active |
| $\hat{D}$ | Landau active, but not Lifshitz active |
| $\bar{\hat{D}}$ | Neither Landau active nor Lifshitz active |

TABLE II. Allowed irreducible representations and allowed subgroups of the two-dimensional space group No. 2: p211. $G_{\vec{k}}=2\ (C_2)$.

| | $^*\vec{k}=\{(\frac{1}{2},0)\}$ $p(2\times1)$ | $^*\vec{k}=\{(0,\frac{1}{2})\}$ $p(1\times2)$ | $^*\vec{k}=\{(\frac{1}{2},\frac{1}{2})\}$ $c(2\times2)$ |
|---|---|---|---|
| $D^j_{G_0}$ | $p211$ | $p211$ | $p211$ |
| $A$ | 1 | 1 | 1 |
| $B$ | 0 | 0 | 0 |

and $l_j=|D^j_{G_0}|$; (4) determine which sets $\{\bar{C}^j_\alpha\}$ will minimize $\Phi$; (5) construct the density function for the system, $\rho(\vec{r})=\rho_0(\vec{r})+\delta\bar{\rho}(\vec{r})$, where $\rho_0(\vec{r})$ is invariant under all symmetry elements $g_0$ in $G_0$, and

$$\delta\bar{\rho}(\vec{r})=\sum_{\alpha=1}^{l_j}\bar{C}^j_\alpha\psi^j_\alpha(\vec{r})\ ,$$

$\psi^j_\alpha(\vec{r})$ are basis functions of $\vec{r}$, which transform as $D^{\alpha=1}_{G_0}$, and the $\bar{C}^j_\alpha$ used in this expression are members of the minimizing set $\{\bar{C}^j_\alpha\}$. The new symmetry group $\bar{G}$ of the system is defined as the maximal set of symmetry elements $\bar{g}$, which is a subset of $G_0$, such that $\bar{g}\,\delta\bar{\rho}(\vec{r})=\delta\bar{\rho}(\vec{r})$. In order to implement this program, it is necessary to expand $\Phi$ in a series of homogeneous $D^j_{G_0}$-invariant polynomials in the set $\{C^j_\alpha\}$, and to carry out the minimization. The Landau procedure is a roundabout method of determining symmetry change.

In order to predict symmetry change directly, the subduction criteria, chain subduction criteria, and chain subduction with multiplicity were introduced.[6] These selection rules are necessary criteria for the "active" irreducible representation $D^j_{G_0}$ to participate in the specified transition from $G_0$ to a subgroup. The order parameters are bases of $D^j_{G_0}$. Thus if a downward-pointing arrow ($\downarrow$) means "subduces" (restricts) and $G_0\supset\bar{G}\supset\bar{G}'$ is a three-chain of maximal subgroups, and if

$$(D^j_{G_0}\downarrow\bar{G})\ni D^{1+}_{\bar{G}}\quad(p\text{ times})$$

and

$$(D^j_{G_0}\downarrow\bar{G}')\ni D^{1+}_{\bar{G}'}\quad(q\text{ times})\ ,$$

then for $p>0$, $G_0\to\bar{G}$ is permitted. For $q=p$, $G_0\to\bar{G}$ is permitted, but $G_0\to\bar{G}'$ does not occur. For $q>p$, $G_0\to\bar{G}$ is permitted, and $G_0\to\bar{G}'$ may occur (possibly as a "multicritical" transition).[7] Since subduction is a necessary criterion, it cannot determine which of the permitted $\bar{G}$, $G'$, . . . , will occur, i.e., give a minimum free energy. However, any subgroup which fails to satisfy the subduction criteria is eliminated.

We have used these direct group-theoretical criteria, or selection rules, to determine all allowed group-subgroup transitions in two dimensions. The subduction selection rules in no way utilize the property that $D^j_{G_0}$ is "active" but could be applied to *any* representation of $G_0$. However, since we choose to work in the general Landau framework, we should first select the active, physically irreducible representations. An immediate problem needs to be confronted, since the "canonical" procedure[4] *requires* that both Landau and Lifshitz criteria be satisfied. A "Landau-active" representation $D^j_{G_0}$ satisfies the stability condition

$$(D^j_{G_0})_{(3)}\not\ni D^{1+}_{G_0}\ ,$$

TABLE III. Allowed irreducible representations and allowed subgroups of the two-dimensional space group No. 3: $p\,1m\,1$. $G_{\vec{k}}=m\ (C_s)$.

| | $^*\vec{k}=\{(\frac{1}{2},0)\}$ $p(2\times1)$ | $^*\vec{k}=\{(0,\frac{1}{2})\}$ $p(1\times2)$ | | $^*\vec{k}=\{(\frac{1}{2},\frac{1}{2})\}$ $c(2\times2)$ |
|---|---|---|---|---|
| $D^j_{G_0}$ | $p\,1m\,1$ | $p\,1m\,1$ | $p\,1g\,1$ | $c\,1m\,1$ |
| $A'$ | 1 | 1 | 0 | 1 |
| $A''$ | 0 | 0 | 1 | 0 |

TABLE IV. Allowed irreducible representations and allowed subgroups of the two-dimensional space group No. 4: $p\,1g\,1$. $G_{\vec{k}}=m\ (C_s)$.

| $*\vec{k}=\{(\frac{1}{2},0)\}$ $D^j_{G_0}$ | $p(2\times1)$ $p\,1g\,1$ |
|---|---|
| $A'$ | 1 |
| $A''$ | 0 |

TABLE V. Allowed irreducible representations and allowed subgroups of the two-dimensional space group No. 5: $c\,1m\,1$. $G_{\vec{k}}=m\ (C_s)$.

| $*\vec{k}=\{(\frac{1}{2},\frac{1}{2})\}$ | $c(2\times2)$ | |
|---|---|---|
| $D^j_{G_0}$ | $p\,1m\,1$ | $p\,1g\,1$ |
| $A'$ | 1 | 0 |
| $A''$ | 0 | 1 |

where $D^{1+}_{G_0}$, as before, is the trivial representation of $G_0$, and the subscript (3) means the symmetrized cube. A Lifshitz-active representation satisfies the homogeneity condition

$$(D^j_{G_0})_{[2]} \not\supset D^v_{G_0}\ ,$$

where $D^v_{G_0}$ is the vector representation of $G_0$, and the subscript [2] means the antisymmetrized square. Both of these rules for deciding "activity" seem to be accepted in three dimensions. But for two-dimensional systems microscopic models have been developed (e.g., three- and four-state Potts model) for which the order parameter violates the Landau condition and yet the transition is continuous.[1] A body of opinion thus holds that the Landau condition should be dropped for all two-dimensional systems. However, the original Landau argument based on stability at the transition seems to have a validity independent of the dimension. We prefer

TABLE VI. Allowed irreducible representations and allowed subgroups of the two-dimensional space group No. 6: $p\,2mm$. $G_{\vec{k}}=2mm\ (C_{2v})$.

| $*\vec{k}=(0,0)$ $D^j_{G_0}$ | $p\,1m\,1$ | $p\,11m$ | $p\,211$ |
|---|---|---|---|
| $A_2$ | 0 | 0 | 1 |
| $B_1$ | 1 | 0 | 0 |
| $B_2$ | 0 | 1 | 0 |

| $*\vec{k}=\{(\frac{1}{2},0)\}$ | | | $p(2\times1)$ | | | |
|---|---|---|---|---|---|---|
| $D^j_{G_0}$ | $p\,2mm$ | $p\,2mg$ | $p\,1m\,1$ | $p\,11m$ | $p\,11g$ | $p\,211$ |
| $A_1$ | 1 | 0 | [1] | [1] | 0 | [1] |
| $A_2$ | 0 | 1 | 0 | 0 | [1] | [1] |
| $B_1$ | 0 | 0 | 1 | 0 | 1 | 0 |
| $B_2$ | 0 | 0 | 0 | 1 | 0 | 0 |

| $*\vec{k}=\{(0,\frac{1}{2})\}$ | | | $p(1\times2)$ | | |
|---|---|---|---|---|---|
| $D^j_{G_0}$ | $p\,2mm$ | $p\,1m\,1$ | $p\,11m$ | $p\,1g\,1$ | $p\,211$ |
| $A_1$ | 1 | [1] | [1] | 0 | [1] |
| $A_2$ | 0 | 0 | 0 | 1 | 1 |
| $B_1$ | 0 | 0 | 0 | 0 | 0 |
| $B_2$ | 0 | 0 | 1 | 1 | 0 |

| $*\vec{k}=\{(\frac{1}{2},\frac{1}{2})\}$ | | $c(2\times2)$ | | |
|---|---|---|---|---|
| $D^j_{G_0}$ | $c\,2mm$ | $c\,1m\,1$ | $c\,11m$ | $p\,211$ |
| $A_1$ | 1 | [1] | [1] | [1] |
| $A_2$ | 0 | 0 | 0 | 1 |
| $B_1$ | 0 | 1 | 0 | 0 |
| $B_2$ | 0 | 0 | 1 | 0 |

TABLE VII. Allowed irreducible representations and allowed subgroups of the two-dimensional space group No. 7: $p2mg$. $G_{\vec{k}}=2mm$ ($C_{2v}$). Tables (e) and (f) are character tables for irreducible ray representations for the indicated wave vectors.

(a) $*\vec{k}=(0,0)$

| $D^j_{G_0}$ | $p11g$ | $p1m1$ | $p211$ |
|---|---|---|---|
| $A_2$ | 0 | 0 | 1 |
| $B_1$ | 0 | 1 | 0 |
| $B_2$ | 1 | 0 | 0 |

(b) $*\vec{k}=\{(0,\frac{1}{2})\}$ $p(1\times 2)$

| $D^j_{G_0}$ | $p2gg$ | $p11g$ | $p1g1$ | $p1m1$ | $p211$ |
|---|---|---|---|---|---|
| $A_1$ | 0 | 1 | 0 | 1 | 1 |
| $A_2$ | 1 | 0 | [1] | 0 | [1] |
| $B_1$ | 0 | 1 | 0 | 1 | 0 |
| $B_2$ | 0 | 0 | 1 | 0 | 0 |

(c) $*\vec{k}=\{(\frac{1}{2},0)\}$ $p(2\times 1)$

| $D^j_{G_0}$ | $p1m1$ | $p211$ |
|---|---|---|
| $\hat{X}$ | 1 | 1 |

(d) $*\vec{k}=\{(\frac{1}{2},\frac{1}{2})\}$ $c(2\times 2)$

| $D^j_{G_0}$ | $p211$ |
|---|---|
| $\hat{S}$ | 1 |

(e) $*\vec{k}=(\frac{1}{2},0)$

| | $E$ | $C_{2z}$ | $\{\sigma_x(\frac{1}{2},0)\}$ | $\{\sigma_y(\frac{1}{2},0)\}$ |
|---|---|---|---|---|
| $X$ | 2 | 0 | 0 | 0 |

(f) $*\vec{k}=(\frac{1}{2},\frac{1}{2})$

| | $E$ | $C_{2z}$ | $\{\sigma_x\mid(\frac{1}{2},0)\}$ | $\{\sigma_y\mid(\frac{1}{2},0)\}$ |
|---|---|---|---|---|
| $S$ | 2 | 0 | 0 | 0 |

not to take a position on this question since we can apply the group-theoretical selection rules to any given representations. Hence we shall indicate those results which pertain to each particular representation and also give their respective activity. In the tables which follow, we denote representations which satisfy *both* the Landau and Lifshitz criteria by $D^j_{G_0}$; those satisfying the Landau, but *not* the Lifshitz, criterion are denoted $\hat{D}^j_{G_0}$; those satisfying the Lifshitz, but *not* the Landau, criterion are denoted by $\bar{D}^j_{G_0}$. Rottman[2] listed LGW Hamiltonians (universality classes) for Lifshitz-active representations only. The first step in our work was to classify representations of each of the 17 planar space groups in this way.

We next require a complete listing of all subgroups for each of these 17 two-dimensional space groups. Such listings exist[8] but not in the most useful form. We restructured the existing listings by taking advantage of a theorem of Hermann.[9] This theorem states that an arbitrary subgroup of a space group is a class-equivalent (same factor group) subgroup of a cell-equivalent (same translation group) subgroup of the original space group. The theorem permits us to prepare a "zellengleich-klassengleich" (Z-K) tree for each space group: The apex of the tree being the parent two-dimensional group and branches being various subgroups. The Z-K tree systematizes the work of subduction and chain subduction. Elsewhere we discussed construction and use of the Z-K tree, and Hermann's theorem in the implementation of the subduction rules. Since it would detract from the continuity of this presentation to give details of the Z-K method, we refer the reader to that paper for details.[10]

## III. RESULTS: ALLOWED TRANSITIONS IN TWO DIMENSIONS

For each nontrivial two-dimensional (2D) space group we shall list the subgroups allowed via second-order phase transitions. In Tables II—XVII irreducible representations (irreps) are labeled with respect to activity as follows (see Table I): $D^j_{G_0}=D$, both Landau and Lifshitz active; $D^j_{G_0}=\bar{D}$, not Landau active, but Lifshitz active; $D^j_{G_0}=\hat{D}$, Landau ac-

TABLE VIII. Allowed irreducible representations and allowed subgroups of the two-dimensional space group No. 8: $p2gg$. $G_{\vec{k}}=2mm$ ($C_{2v}$). Tables (c), (f), and (g) are character tables for irreducible ray representations for the indicated wave vectors.

(a) $*\vec{k}=(0,0)$

| $D^j_{G_0}$ | $p1g1$ | $p11g$ | $p211$ |
|---|---|---|---|
| $A_2$ | 1 | 0 | 0 |
| $B_1$ | 0 | 1 | 0 |
| $B_2$ | 0 | 0 | 1 |

(b) $*\vec{k}=\{(\frac{1}{2},\frac{1}{2})\}$ $c(2\times2)$

| $D^j_{G_0}$ | $p211$ |
|---|---|
| $S_1$ | 2 |
| $S_2$ | 0 |

| (c) $*\vec{k}=(\frac{1}{2},\frac{1}{2})$ | $E$ | $C_{2z}$ | $\{\sigma_x \mid (\frac{1}{2},\frac{1}{2})\}$ | $\{\sigma_y \mid (\frac{1}{2},\frac{1}{2})\}$ |
|---|---|---|---|---|
| $S_1$ | 2 | 2 | 0 | 0 |
| $S_2$ | 2 | $-2$ | 0 | 0 |

(d) $*\vec{k}=\{(\frac{1}{2},0)\}$ $p(2\times1)$

| $D^j_{G_0}$ | $p1g1$ | $p211$ |
|---|---|---|
| $\hat{X}$ | 1 | 1 |

(e) $*\vec{k}=\{(0,\frac{1}{2})\}$ $p(1\times2)$

| $D^j_{G_0}$ | $p11g$ | $p211$ |
|---|---|---|
| $\hat{Y}$ | 1 | 1 |

| (f) $*\vec{k}=(\frac{1}{2},0)$ | $E$ | $C_{2z}$ | $\{\sigma_x \mid (\frac{1}{2},\frac{1}{2})\}$ | $\{\sigma_y \mid (\frac{1}{2},\frac{1}{2})\}$ |
|---|---|---|---|---|
| $X$ | 2 | 0 | 0 | 0 |

| (g) $*\vec{k}=(0,\frac{1}{2})$ | $E$ | $C_{2z}$ | $\{\sigma_x \mid (\frac{1}{2},\frac{1}{2})\}$ | $\{\sigma_y \mid (\frac{1}{2},\frac{1}{2})\}$ |
|---|---|---|---|---|
| $Y$ | 2 | 0 | 0 | 0 |

tive, but not Lifshitz active; $D^j_{G_0}=\overline{\hat{D}}$, neither Landau active nor Lifshitz active.

The point group of the $\vec{k}$ vector $G_{\vec{k}}$, the active star of $\vec{k}$ ($*\vec{k}$), and superlattices $p(m\times n)$, $p(\sqrt{m}\times\sqrt{m})R\theta$, etc., are given in the first row of each table. In those cases for which characters of elements in $G_k$ cannot be obtained from the ordinary point-group tables, we give the explicit character in a small table appended and we follow the notation of Cracknell.[8] Integer entries in the column below each subgroup of $G_0$ are the subduction frequencies calculated by the group-theoretical methods given in the text and in Refs. 6 and 7. The notation used is as follows: $n=1,2,\ldots$, indicates an allowed subgroup; $n=[1]$ or $[2]$ indicates that a particular subgroup is eliminated via the chain criteria.

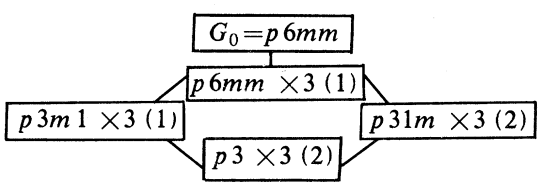
As an example of the application of the chain criteria in eliminating certain subgroups we consider Table XVII space group No. 17, $p6mm$ $[p(\sqrt{3}\times\sqrt{3})R\,30°]$ with $*\vec{k}=\{(-\frac{1}{3},\frac{2}{3}),(\frac{1}{3},\frac{1}{3})\}$ and $D^j_{G_0}=\overline{A}_1$. This irreducible representation is not Landau active, but is Lifshitz active, and in three dimensions (3D), it would not be considered at all.

We have the following two chains of subgroups and subduction frequencies $(p,q,\ldots)$ from the tables:

TABLE IX. Allowed irreducible representations and allowed subgroups of the two-dimensional space group No. 9: $c2mm$.

| $G_{\vec{k}}=2mm\ (C_{2v})$ | | | | $G_{\vec{k}}=2(C_2)$ | | |
|---|---|---|---|---|---|---|
| $*\vec{k}=(0,0)$ | | | | $*\vec{k}=\{(\frac{1}{2},0),(0,\frac{1}{2})\}$ | $p(2\times2)$ | $p(2\times1)$ |
| $D^j_{G_0}$ | $c1m1$ | $c11m$ | $p211$ | $D^j_{G_0}$ | $c2mm$ | $p211$ |
| $A_2$ | 0 | 0 | 1 | $A$ | 1 | 1 |
| $B_1$ | 1 | 0 | 0 | $B$ | 0 | 0 |
| $B_2$ | 0 | 1 | 0 | | | |

| $*\vec{k}=\{(\frac{1}{2},\frac{1}{2})\}$ | $G_{\vec{k}}=2mm\ (C_{2v})$ $c(2\times2)$ | | | | | |
|---|---|---|---|---|---|---|
| $D^j_{G_0}$ | $p2mm$ | $p2mg$ | $p2gg$ | $p1m1$, $p1g1$ | $p11m$, $p11g$ | $p211$ |
| $A_1$ | 1 | 0 | 0 | [1] | [1] | [1] |
| $A_2$ | 0 | 0 | 1 | 0 | 0 | [1] |
| $B_1$ | 0 | 0 | 0 | 1 | 0 | 0 |
| $B_2$ | 0 | 1 | 0 | 0 | [1] | 0 |




The threefold increase in cell area is indicated by $\times3$. The corresponding subduction frequencies are enclosed in parentheses.

Applying the chain subduction criteria to the three-chain on the left, we see that $p6mm \rightarrow p3m1$ is not allowed. In the three-chain on the right, $p6mm \rightarrow p3$ is not allowed. This leaves $p6mm \rightarrow p6mm \times 3$ and $p6mm \rightarrow p31m\ \times3$. Hence the entries in Table XVIII are the following:

| $p3m1$ | $p3$ | $p31m$ | $p6mm$ |
|---|---|---|---|
| [1] | [2] | 2 | 1 |

In brief, for a subgroup to be allowed, it must survive elimination in *every* chain in which it occurs. If it is eliminated in any chain, it is not allowed for the second-order phase transitions.[11]

Recently, a listing of symmetry breaking by second-order phase transitions in two dimensions was given by Ipatova *et al.*[12] That work considered only superlattice formation, i.e., breaking only the translational symmetry. Our work extends and completes their results to include all space-group to

TABLE X. Allowed irreducible representation and allowed subgroups of the two-dimensional space group No. 10: $p4$.

| $G_{\vec{k}}=4\ (C_4)$ | | $G_{\vec{k}}=2\ (C_2)$ | | $G_{\vec{k}}=4\ (C_4)$ | | |
|---|---|---|---|---|---|---|
| $*\vec{k}=(0,0)$ | | $*\vec{k}=\{(\frac{1}{2},0)(0,\frac{1}{2})\}$ | $p(2\times1)$ | $*\vec{k}=\{(\frac{1}{2},\frac{1}{2})\}$ | $c(2\times2)$ | |
| $D^j_{G_0}$ | $p211$ | $D^j_{G_0}$ | $p211$ | $D^j_{G_0}$ | $p4$ | $p211$ |
| $B$ | 1 | $A$ | 1 | $A$ | 1 | [1] |
| | | | | $B$ | 0 | 1 |
| $\hat{E}$ | 0 | $B$ | 0 | $E$ | 0 | 0 |

TABLE XI. Allowed irreducible representations and allowed subgroups of the two-dimensional space group No. 11: *p4mm*.

| $*\vec{k}=(0,0)$ | $G_{\vec{k}}=4mm\ (C_{4v})$ | | | | |
|---|---|---|---|---|---|
| $D^{j}_{G_0}$ | $c2mm$ / $p2mm$ | $p4$ | $c1m1$ / $c11m$ | $p1m1$ / $p11m$ | $p211$ |
| $A_2$ | 0 | 1 | 0 | 0 | [1] |
| $B_1$ | 1 | 0 | [1] | [1] | [1] |
| $B_2$ | 0 | 0 | 0 | 0 | 1 |
| $E$ | 0 | 0 | 1 | 1 | 0 |

| $*\vec{k}=\{(\frac{1}{2},0),(0,\frac{1}{2})\}$ | $G_{\vec{k}}=2mm\ (C_{2v})$ $p(2\times1)$ | | | | | | $p(2\times2)$ |
|---|---|---|---|---|---|---|---|
| $D^{j}_{G_0}$ | $p2mm$ | $p2mg$ | $p11g$ | $p1m1$ | $p11m$ | $p211$ | $p2gg$ |
| $A_1$ | 1 | 0 | 0 | [1] | [1] | [1] | 0 |
| $A_2$ | 0 | 1 | [1] | 0 | 0 | [1] | 2 |
| $B_1$ | 0 | 0 | 1 | 0 | 0 | 0 | 0 |
| $B_2$ | 0 | 0 | 0 | 0 | 1 | 0 | 0 |

| $*\vec{k}=\{(\frac{1}{2},\frac{1}{2})\}$ | $G_{\vec{k}}=4mm\ (C_{4v})$ $c(2\times2)$ or $p(\sqrt{2}\times\sqrt{2})R45°$ | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| $D^{j}_{G_0}$ | $p4mm$ | $p4gm$ | $p4$ | $c2mm$ | $p2mg$ | $p2gg$ | $p1m1$ / $p11m$ | $p1g1$ | $c1m1$, $c11m$, $p11g$ | $p211$ |
| $A_1$ | 1 | 0 | [1] | [1] | 0 | 0 | [1] | 0 | [1] | [1] |
| $A_2$ | 0 | 1 | [1] | 0 | 0 | [1] | 0 | [1] | 0 | [1] |
| $B_1$ | 0 | 0 | 0 | 0 | 0 | 1 | 0 | [1] | 0 | [1] |
| $B_2$ | 0 | 0 | 0 | 1 | 0 | 0 | [1] | 0 | [1] | [1] |
| $E$ | 0 | 0 | 0 | 1 | 1 | 0 | [1] | [1] | [1] | 0 |

space-group transitions from one of the 17 plane groups to another, allowing both rotational- and translational-symmetry breaking.

There seem to be few experimental determinations of symmetry change and order of the phase transition for surfaces with which to compare our results. The work of Felter, Barker, and Estrup,[13] and of Debe and King,[14] which reports a second-order phase transition on the W{100} surface, agrees with our predictions as given in Table XI for the two-dimensional space group *p4mm*, with the star of $\vec{k}$, $*\vec{k}=\{(\frac{1}{2},\frac{1}{2})\}$. An allowed second-order transition can occur to $p(\sqrt{2}\times\sqrt{2})R45°$, with the two-dimensional space group *p2mg*, using the irreducible representation $D^{j}_{G_0}=E$.

In the Appendix we have investigated this transition using the standard Landau procedure[4] by minimizing a fourth-degree polynomial. It is easy to verify that the subduction methods are more convenient and that they give the same information.

After our work was completed we became aware of some interesting work on structural phase transitions for the admolecular system of $CH_4$ on (0001) graphite by Marx and Wasserman.[5] They developed a Landau theory[4] for the transition. Marx's prediction by minimization of a Landau polynomial of a lower-symmetry group *c2mm* for $*\vec{k}=\{(\frac{1}{2},0),(0,-\frac{1}{2}),(-\frac{1}{2}\frac{1}{2})\}$ agrees with our direct group-theory results, for $G_0=p6mm$ and for $D^{j}_{G_0}=A_2$ ($A'_2$ in Ref. 15). We find the subgroup $c2mm$ allowed, with subduction frequency $n=1$ and a hexagonal $p(2\times2)$ superlattice. The primitive vectors of a deformed hexagonal lattice (with $\alpha\neq120°$) define a centered rectangular lattice. The space-group symmetry would be $c2mm$ as given in Marx's (Ref. 15) Fig. 4 and the accompanying text.

In the final summary table (Table XVIII) we list all the results of our work in a convenient form (see the table of subgroups of plane groups in Coxeter and Moser[3]). Row heading is the parent group $G_0$,

TABLE XII. Allowed irreducible representations and allowed subgroups of the two-dimensional space group No. 12: $p4gm$. Tables (c) and (e) are character tables for irreducible ray representations for the indicated wave vectors.

| (a) $^*\vec{k}=(0,0)$ | $G_{\vec{k}}=4mm\ (C_{4v})$ | | | | | |
|---|---|---|---|---|---|---|
| $D^j_{G_0}$ | $p4$ | $c2mm$ | $p2gg$ | $c1m1$, $c11m$ | $p1g1$, $p11g$ | $p211$ |
| $A_2$ | 1 | 0 | 0 | 0 | 0 | [1] |
| $B_1$ | 0 | 1 | 1 | [1] | [1] | [1] |
| $B_2$ | 0 | 0 | 0 | 0 | 0 | 1 |
| $E$ | 0 | 0 | 0 | 1 | 1 | 0 |

| (b) $^*\vec{k}=\{(\frac{1}{2},0),(0,\frac{1}{2})\}$ | $G_{\vec{k}}=2mm\ (C_{2v})$, $p(2\times1)$ | |
|---|---|---|
| $D^j_{G_0}$ | $p1g1$ | $p211$ |
| $\hat{X}$ | 1 | 1 |

| (c) $^*\vec{k}=(\frac{1}{2},0)$ | $E$ | $C_{2z}$ | $\{\sigma_x\mid(\frac{1}{2},\frac{1}{2})\}$ | $\{\sigma_y\mid(\frac{1}{2},\frac{1}{2})\}$ |
|---|---|---|---|---|
| $X$ | 2 | 0 | 0 | 0 |

| (d) $^*\vec{k}=\{(\frac{1}{2},\frac{1}{2})\}$ | $G_{\vec{k}}=4mm\ (C_{4v})$, $c(2\times2)$ or $p(\sqrt{2}\times\sqrt{2})R45^\circ$ | | | | | | |
|---|---|---|---|---|---|---|---|
| $D^j_{G_0}$ | $p4$ | $p2mm$, $c2mm$ | $p2mg$ | $p2gg$ | $p1g1$, $p11g$ | $p1m1$, $p11m$ | $p211$ |
| $M'$ | 1 | 0 | 1 | 0 | [1] | [1] | 2 |
| $M''$ | 0 | 2 | 0 | 2 | [2] | [2] | 0 |

| (e) $^*\vec{k}=(\frac{1}{2},\frac{1}{2})$ | $E$ | $C_{4z}^{\pm}$ | $C_{2z}$ | $\{\sigma_x\mid(\frac{1}{2},\frac{1}{2})\}$ | $\{\sigma_y\mid(\frac{1}{2},\frac{1}{2})\}$ | $\{\sigma_{da}\mid(\frac{1}{2},\frac{1}{2})\}$ | $\sigma_{db}\{(\frac{1}{2},\frac{1}{2})\}$ |
|---|---|---|---|---|---|---|---|
| $M'$ | 2 | 0 | 2 | 0 | 0 | 0 | 0 |
| $M''$ | 4 | 0 | $-4$ | 0 | 0 | 0 | 0 |

with all allowed subgroups $\bar{G}$ listed horizontally.[16] The caption to the table indicates the factor of increase of cell area (e.g, $\times2$, etc.), and is a key to the orientation of the subgroup *vis-à-vis* the parent. Subgroup labels $\bar{G}$ are the column headings. In order to determine the active order parameter in each case, one should go back to the detailed Tables II—XVII for the two-dimensional space groups Nos. 2—17 given earlier.

TABLE XIII. Allowed irreducible representations and allowed subgroups of the two-dimensional space group No. 13: $p3$. $G_{\vec{k}}=3\ (C_3)$.

| $^*\vec{k}=\{(-\frac{1}{3},\frac{2}{3})\}$ | $p(\sqrt{3}\times\sqrt{3})R30^\circ$ |
|---|---|
| $D^j_{G_0}$ | $p3$ |
| $\bar{A}$ | 1 |
| $\bar{E}$ | 0 |

TABLE XIV. Allowed irreducible representations and allowed subgroups of the two-dimensional space group No. 14: $p3m1$.

| $*\vec{k}=(0,0)$ | $G_{\vec{k}}=3m$ ($C_{3v}$) | |
|---|---|---|
| $D^j_{G_0}$ | $p3$ | $c1m1$ |
| $\underline{A_2}$ | 1 | 0 |
| $\hat{E}$ | 0 | 1 |

| $*\vec{k}=\{(\frac{1}{2},0),(0,-\frac{1}{2}),(-\frac{1}{2},\frac{1}{2})\}$ | $G_k=m$ ($C_s$) $c(2\times2)$ | | $p(2\times2)$ | $*\vec{k}=\{(-\frac{1}{3},\frac{2}{3}),(\frac{1}{3},\frac{1}{3})\}$ | $G_{\vec{k}}=3$ ($C_3$) $p(\sqrt{3}\times\sqrt{3})R\,30°$ | | |
|---|---|---|---|---|---|---|---|
| $D^j_{G_0}$ | $p1m1$ | $p1g1$ | $p3m1$ | $D^j_{G_0}$ | $p31m$ | $p3$ | $c1m1$ |
| $\bar{A}'$ | 1 | 1 | 1 | $\bar{A}$ | 1 | 2 | [1] |
| $\bar{A}''$ | 0 | 0 | 0 | $\bar{\hat{E}}$ | 0 | 0 | 2 |

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## APPENDIX

In this appendix we analyze the transition $p4mm \rightarrow p2mg$ $[p(\sqrt{2}\times\sqrt{2})R\,45°]$ using conventional Landau theory.[4] Since $G_0=p4mm$ is a symmorphic space group, the representation of the little group of the $\vec{k}$ vector ($G_{\vec{k}}$) at $\vec{K}=(\frac{1}{2},\frac{1}{2})$ is the same as that of the point group of the $\vec{k}$ vector $\bar{G}_k=4mmm$ ($C_{4v}$). We have the following matrices (Bradley and Cracknell[17]) for the two-dimensional representation at $\vec{k}=(\frac{1}{2},\frac{1}{2})$. The group elements are denoted by $g$, the matrices by $D(g)$:

TABLE XV. Allowed irreducible representations and allowed subgroups of the two-dimensional space group No. 15: $p31m$.

| $*\vec{k}=(0,0)$ | $G_{\vec{k}}=3m$ ($C_{3v}$) | | $*\vec{k}=\{(\frac{1}{2},0),(0,-\frac{1}{2}),(-\frac{1}{2},\frac{1}{2})\}$ | $G_{\vec{k}}=m$ ($C_s$) $p(2\times2)$ | $c(2\times2)$ | | $*\vec{k}=\{(-\frac{1}{3},\frac{2}{3})\}$ | $G_{\vec{k}}=3m$ ($C_{3v}$) $p(\sqrt{3}\times\sqrt{3})R\,30°$ | | |
|---|---|---|---|---|---|---|---|---|---|---|
| $D^j_{G_0}$ | $p3$ | $c11m$ | $D^j_{G_0}$ | $p31m$ | $p11m$ | $p11g$ | $D^j_{G_0}$ | $p3m1$ | $p3$ | $c11m$ |
| $\underline{A_2}$ | 1 | 0 | $\bar{A}'$ | 1 | 1 | 0 | $A_1$ | 1 | [1] | [1] |
| $\hat{E}$ | 0 | 1 | $\bar{A}''$ | 0 | 0 | 1 | $A_2$ | 0 | 1 | 0 |
| | | | | | | | $E$ | 0 | 0 | 1 |

TABLE XVI. Allowed irreducible representations and allowed subgroups of the two-dimensional space-group No. 16: *p*6.

| $G_{\vec{k}}=6$ $(C_6)$ | | | $G_{\vec{k}}=2$ $(C_2)$ | | | $G_{\vec{k}}=3$ $(C_3)$ | |
|---|---|---|---|---|---|---|---|
| $*\vec{k}=(0,0)$ | | | $*\vec{k}=\{(\frac{1}{2},0),(0,-\frac{1}{2}),(-\frac{1}{2},\frac{1}{2})\}$ | $p(2\times 2)$ | $c(2\times 2)$ | $*\vec{k}=\{(-\frac{1}{3},\frac{2}{3}),(\frac{1}{3},\frac{1}{3})\}$ | $p(\sqrt{3}\times\sqrt{3})R\,30°$ |
| $D^j_{G_0}$ | $p3$ | $p211$ | $D^j_{G_0}$ | $p6$ | $p211$ | $D^j_{G_0}$ | $p3$ |
| $\underline{B}$ | 1 | 0 | $\bar{A}$ | 1 | 1 | $\bar{A}$ | 2 |
| $\underline{\hat{E}}_1$ | 0 | 2 | $B$ | 0 | 0 | $\underline{\hat{E}}$ | 0 |
| $\hat{E}_2$ | 0 | 0 | | | | | |

| $g$ | $E$ | $C_{4z}^+$ | $C_{4z}^-$ | $C_{2z}$ | $\sigma_x$ | $\sigma_y$ | $\sigma_{da}$ | $\sigma_{db}$ |
|---|---|---|---|---|---|---|---|---|
| $D(g)$ | $\begin{bmatrix}1&0\\0&1\end{bmatrix}$ | $\begin{bmatrix}0&-1\\1&0\end{bmatrix}$ | $\begin{bmatrix}0&1\\-1&0\end{bmatrix}$ | $\begin{bmatrix}-1&0\\0&-1\end{bmatrix}$ | $\begin{bmatrix}-1&0\\0&1\end{bmatrix}$ | $\begin{bmatrix}1&0\\0&-1\end{bmatrix}$ | $\begin{bmatrix}0&-1\\-1&0\end{bmatrix}$ | $\begin{bmatrix}0&1\\1&0\end{bmatrix}$ |

### 1. Free energy

It is easy to construct the invariant Landau polynomial for the free energy $\Phi$. Up to fourth degree,

$$\Phi=A'(c_1^2+c_2^2)+B_1'(c_1^4+c_2^4)+B_2'c_1^2c_2^2+\cdots .$$

Letting $c_1=\rho\cos\theta$ and $c_2=\rho\sin\theta$,

$$\Phi=\frac{A}{2}\rho^2+\frac{B_1}{4}\rho^4+\frac{B_2}{4}\rho^4\cos4\theta .$$

Extremizing gives $4\theta=n\pi$, $n=0,1,2,\ldots$, and

$$\rho\neq 0,\ \ \rho^2=\frac{-A}{B_1+B_2\cos4\theta}=\frac{-A}{B_1\pm B_2} .$$

Possible solutions for the order parameters are the following:

TABLE XVII. Allowed irreducible representations and allowed subgroups of the two-dimensional space group No. 17: *p*6*mm*.

| | $G_{\vec{k}}=6mm$ $(C_{6v})$ | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| $*\vec{k}=(0,0)$ | | | | | | | | |
| $D^j_{G_0}$ | $p6$ | $p3m1$ | $p31m$ | $c2mm$ | $p3$ | $c1m1$ | $c11m$ | $p211$ |
| $A_2$ | 1 | 0 | 0 | 0 | [1] | 0 | 0 | [1] |
| $B_1$ | 0 | 1 | 0 | 0 | [1] | [1] | 0 | 0 |
| $B_2$ | 0 | 0 | 1 | 0 | [1] | 0 | [1] | 0 |
| $\underline{\hat{E}}_1$ | 0 | 0 | 0 | 0 | 0 | 1 | 1 | 0 |
| $\hat{E}_2$ | 0 | 0 | 0 | 1 | 0 | [1] | [1] | 2 |

| | $G_{\vec{k}}=3m$ $(C_{3v})$ | | | | | |
|---|---|---|---|---|---|---|
| $*\vec{k}=\{(-\frac{1}{3},\frac{2}{3}),(\frac{1}{3},\frac{1}{3})\}$ | | | $p(\sqrt{3}\times\sqrt{3})R\,30°$ | | | |
| $D^j_{G_0}$ | $p6mm$ | $p3m1$ | $p31m$ | $p3$ | $c1m1$ | $c11m$ |
| $\bar{A}_1$ | 1 | [1] | 2 | [2] | [1] | [2] |
| $\underline{A}_2$ | 0 | 1 | 0 | 2 | [1] | 0 |
| $\underline{\hat{E}}$ | 0 | 0 | 0 | 0 | 2 | 2 |

TABLE XVII. (*Continued.*)

| $^*\vec{k}=\{(\frac{1}{2},0),(0,-\frac{1}{2}),(-\frac{1}{2},\frac{1}{2})\}$ | | | $G_{\vec{k}}=2mm\ (C_{2v})$ | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | $p(2\times2)$ | | | | | | | $c(2\times2)$ | | | | |
| $D^j_{G_0}$ | $p6mm$ | $p6$ | $p3m1$ | $p31m$ | $c2mm$ | $p2mm$ | $p2mg$ | $p2gg$ | $p1m1$ | $p11m$ | $p1g1$ | $p11g$ | $p211$ |
| $\bar{A}_1$ | 1 | [1] | [1] | [1] | 2 | 1 | 0 | 0 | [1] | [1] | 0 | 0 | [1] |
| $\bar{A}_2$ | 0 | 1 | 0 | 0 | 1 | 0 | 0 | 1 | 0 | 0 | [1] | [1] | [1] |
| $B_1$ | 0 | 0 | 0 | 1 | 0 | 0 | 0 | 0 | 0 | 1 | 1 | 0 | 0 |
| $B_2$ | 0 | 0 | 1 | 0 | 0 | 0 | 1 | 0 | [1] | 0 | 0 | [1] | 0 |

TABLE XVIII. Summary table. Allowed subgroups in continuous phase transitions for the plane space groups. Parent group $G_0$ is the row heading. Column heading is the subgroup $\bar{G}$. Key to symbols in the table (orientation of subgroup *vis-à-vis* parent group). Integer $n=1,2,3,4$ is $n$-fold increase in cell size: $2^a$, $p(2\times1)$; $2^b$, $p(1\times2)$; $2^c$, $c(2\times2)$; $2^d$, $p(\sqrt{2}\times\sqrt{2})R\,45°$; 3, $p(\sqrt{3}\times\sqrt{3})R\,30°$; 4, $p(2\times2)$.

| | $p2$ | $pm$ | $pg$ | $cm$ | $p2mm$ | $p2mg$ | $p2gg$ | $c2mm$ | $p4$ | $p4mm$ | $p4gm$ | $p3$ | $p3m1$ | $p31m$ | $p6$ | $p6mm$ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $p2$ | $2^{a,b,c}$ | | | | | | | | | | | | | | | |
| $pm$ | | $2^{a,b}$ | $2^b$ | $2^c$ | | | | | | | | | | | | |
| $pg$ | | | $2^a$ | | | | | | | | | | | | | |
| $cm$ | | $2^c$ | $2^c$ | | | | | | | | | | | | | |
| $p2mm$ | $1,2^{a,b,c}$ | $1,2^{a,b}$ | $2^{a,b}$ | $2^c$ | $2^{a,b}$ | $2^a$ | | $2^c$ | | | | | | | | |
| $p2mg$ | $1,2^{a,b,c}$ | $1,2^{a,b}$ | $1,2^b$ | | | | $2^b$ | | | | | | | | | |
| $p2gg$ | $1,2^{a,b,c}$ | | $1,2^{a,b}$ | | | | | | | | | | | | | |
| $c2mm$ | $1,2^a$ | $2^c$ | $2^c$ | 1 | $2^c$ | $2^c$ | $2^c$ | 4 | | | | | | | | |
| $p4$ | $1,2^{a,c}$ | | | | | | | | $2^c$ | | | | | | | |
| $p4mm$ | 1 | $1,2^a$ | $2^a$ | 1 | $1,2^a$ | $2^{a,d}$ | $2^d,4$ | $1,2^d$ | 1 | $2^d$ | $2^d$ | | | | | |
| $p4gm$ | $1,2^{a,d}$ | | $1,2^a$ | 1 | $2^d$ | $2^d$ | $1,2^d$ | $1,2^d$ | $1,2^d$ | | | | | | | |
| $p3$ | | | | | | | | | | | | 3 | | | | |
| $p3m1$ | | $2^c$ | $2^c$ | 1,3 | | | | | | | | 1,3 | 4 | 3 | | |
| $p31m$ | | $2^c$ | $2^c$ | 1,3 | | | | | | | | 1,3 | 3 | 4 | | |
| $p6$ | $1,2^c$ | | | | | | | | | | | 1,3 | | | 4 | |
| $p6mm$ | 1 | $2^c$ | $2^c$ | 1,3 | $2^c$ | $2^c$ | $2^c$ | 1,4 | | | | 3 | 1,3,4 | 1,3,4 | 1,4 | 3,4 |

$$c_1 \neq 0,\ c_2 = 0, \quad \text{for } \theta = 0 ,$$
$$c_1 = 0,\ c_2 \neq 0, \quad \text{for } \theta = \pi/2 ,$$
$$c_1 = +c_2, \quad \text{for } \theta = \pi/4 ,$$
$$c_1 = -c_2, \quad \text{for } \theta = 3\pi/4 .$$

To find the minima, we evaluate $\partial^2\Phi/\partial\theta^2$ and $\partial^2\Phi/\partial\rho^2$, and $\Phi$:

$$\frac{\partial^2\Phi}{\partial\theta^2} = -4B_2\rho^4\cos 4\theta = -4(\pm B_2)\rho^4 ,$$

so that

$$\frac{\partial^2\Phi}{\partial\theta^2} > 0 \quad \text{for} \quad \begin{cases} B_2 < 0 & c_1 \neq 0,\ c_2 = 0 \ \ (\theta = 0) \\ & c_1 = 0,\ c_1 \neq 0 \ \ (\theta = \pi/2) \\ B_2 > 0 & c_1 = c_2 \ \ (\theta = \pi/4) \\ & c_1 = -c_2 \ \ (\theta = 3\pi/4) \end{cases}$$

$$\frac{\partial^2\Phi}{2\rho^2} = A - \frac{3A}{(B_1 \pm B_2)}(B_1 \pm B_2) = -2A ,$$

so that

$$\partial^2\Phi/\partial\rho^2 > 0 \text{ for } A < 0 ,$$

$$\Phi = \frac{-A^2}{4(B_1 \pm B_2)} ,$$

so that $\Phi < 0$ for $B_1 > 0$ and $B_1 > |B_2|$.

### 2. Lower-symmetry space group

Having established absolute minima for the above roots, we now examine the density function $\delta\rho = c_1\psi_1 + c_2\psi_2$. For $c_1 = +c_2$, $c_1(\psi_1 + \psi_2)$ transforms under the elements of $p4mm$ as

| $p4mm$ | $E$ | $C_{4z}^{+}$ | $C_{4z}^{-}$ | $C_{2z}$ | $\sigma_x$ | $\sigma_y$ | $\sigma_{da}$ | $\sigma_{db}$ |
|---|---|---|---|---|---|---|---|---|
| $\psi_1+\psi_2$ | $\psi_1+\psi_2$ | $-\psi_2+\psi_1$ | $\psi_2-\psi_1$ | $-(\psi_1+\psi_2)$ | $-\psi_1+\psi_2$ | $\psi_1-\psi_2$ | $-(\psi_2+\psi_1)$ | $\psi_2+\psi_1$ |

$\delta\rho$ is invariant under the action of the elements $\{E|(0,0)\}$, $\{C_{2z}|\vec{t}_i\}$, $\{\sigma_{da}|\vec{t}_i\}$, and $\{\sigma_{db}|(0,0)\}$, where $i = 1,2$; $\vec{t}_1 = (1,0)$ and $\vec{t}_2 = (0,1)$. Shifting the origin by $\vec{t}_0 = (\frac{1}{2},0)$ to agree with Bradley and Cracknell,[17] we finally obtain $\{E|(0,0)\}$, $\{C_{2z}|(0,0)\}$, $\{\sigma_{da}|\vec{\tau}_2'/2\}$, and $\{\sigma_{db}|-\vec{\tau}_2'/2\}$, where the new lattice translations are the following:

$$\vec{\tau}_1' = (\vec{t}_1 + \vec{t}_2) ,$$

$$\vec{\tau}_2' = (\vec{t}_1 - \vec{t}_2) .$$

We identify the space group as $p2mg$ $[p(\sqrt{2}\times\sqrt{2})R45°]$. In a similar way $c_1 \neq 0$, $c_2 = 0$ leads to the transition $p4mm \rightarrow c2mm$ with a volume increase of a factor of 2. The other roots $c_1 = -c_2$ and $c_1 = 0$, $c_2 \neq 0$ are related to the above by symmetry and give the same transitions.


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