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Phasons and Amplitudons of the OSICDW of $2H\text{-}TaSe_2$ in McMillan-Landau Theory

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Abstract Analytical computations are performed to obtain the orthorhombic stripe incommensurate charge-density-wave (OSICDW) phase eigenmode frequencies of the transition metal dichalcogenide $2H\text{-}TaSe_2$, using the McMillan-Landau theory of phase transitions. The amplitudons and phasons have been obtained for a particular direction, where it is possible to obtain simpler expression for them.

1. INTRODUCTION

In the precedent article[1] we have computed, from the McMillan-Landau free energy potential, the eigenmode frequencies for the hexagonal incommensurate CDW phase of $2H\text{-}TaSe_2$.

Now we compute the same type of excitations (amplitudons and phasons) for the case of the orthorhombic stripe incommensurate charge-density-wave phase of this compound, using the same approach, which has been very convenient[2], for the case of incommensurate superlattices.

The outline of this note is the following: first, we summarize some experimental evidences for this phase, second, we write the new free energy potential computing the order-parameter fluctuation modes for this structure and finally we discuss the analytical results.

2. THEORY

It is well established that the transition metal dichalcogenide $2H\text{-}TaSe_2$ exhibits an orthorhombic 'striped' incommesurate phase between $90^{\circ}K$ and $112^{\circ}K$. Fleming *et al*[3] detected a large hysteresis effect in which $T_{CI} = 85^{\circ}K$ on cooling and $T_{CI} = 93^{\circ}K$ on warming, where T_{CI} is the commensurate-incornmensurate transition temperature. They also observed that on warming between $93^{\circ}K$ and $112^{\circ}K$, the hexagonal symmetry of the triple - $\vec{q}_3$ charge-density-wave vectors $|\vec{q}_1| = |\vec{q}_2| = |\vec{q}_3|$ was broken in

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a new triple - $\vec{q}_j$ CDW, where one of the three $\vec{q}_i$ remains commensurate and the other two vectors become incommensurate. Diffraction satellites have identified the new phase as having a striped geometry[4]. This experimental work corroborated the original results of Steinitz and Genossar[5]. Other authors[6] have confirmed that within the region of the striped phase, parallel commensurate stripes separated by narrow domain walls occur and that above 112°K a first-order phase change occurs and the stripe-domain structure transforms to a domain structure with hexagonal symmetry.

In the present calculations we are restricted to computing order-parameter fluctuation modes for the orthorhombic stripe incommensurate structure in $2H\text{-}TaSe_2$, which is characterized by the space group C_{2V}-2mm, using the McMillan-Landau free energy potential taking into account intralayer contributions.

Following Fleming et al[3] we define, in this phase, the three $\vec{q}_j$ - CDW vectors:

$$\vec{q}_1 = \frac{1}{3}\vec{K}_1 \ , \quad \vec{q}_2 = \frac{1}{3}\vec{K}_2 - \vec{k} \ , \quad \vec{q}_3 = \frac{1}{3}\vec{K}_3 + \vec{k} \ ; \qquad (1)$$

$\vec{q}_1$ being the commensurate wave-vector and $\vec{q}_2$ and $\vec{q}_3$ the incommensurate ones.

Using the same procedure as in ref, 1, with $\phi_1 \neq \phi_2 = \phi_3$, after some algebraic manipulations the new McMillan-Landau free energy can be written as

$$V = \frac{1}{2}\int d^2r \ \{\sum_{j=1}^{3} [a_0|\phi_j|^2 + \frac{1}{4}3c_0|\phi_j|^4 + 2e_0|\vec{Q}_j.\{\vec{\nabla} + i[\frac{1}{3}\vec{K}_j - \vec{Q}_j - (-1)^j\vec{k}]\}\phi_j|^2]$$

$$+ 2f_0[|\vec{Q}_1\times\vec{\nabla}\phi_1|^2 + \sum_{j=2}^{3}|\vec{Q}_j\times[\vec{\nabla} - (-1)^j i\vec{k}]\phi_j|^2] - \frac{1}{2}\,\mathrm{Re}\,(b_1\phi_1^3) \qquad (2)$$

$$+ (3c_0+2d_0)(|\phi_1\phi_2|^2 + |\phi_2\phi_3|^2 + |\phi_3\phi_1|^2) - \frac{1}{2}3b_0(\phi_1\phi_2\phi_3 + \phi_1^*\phi_2^*\phi_3^*)\} \ ,$$

where we obtain a cubic 'lock-in' contribution only in the commensurate $\vec{q}_1$-direction. The stripes are parallel to this direction. The phenomenological parameters a_0, c_0, d_0, ... and different vectors in eq. (2) have already been defined in ref, 1.

In order to compute the eigenmode frequencies, we expand (2) in powers of ϕ_{jq} keeping only second-order terms and find

$$V = V_0 + \sum_q \{ \sum_j [(\frac{1}{2}a_0 + \frac{1}{2}\, 3c_0\phi_j^2 + \Lambda_{jj})\phi^*_{jq}\phi_{jq} + \frac{1}{8}\, 3c_0\phi_j^2(\phi^*_{jq}\phi^*_{j-q} - \mathbb{M}_{jq}\mathbb{M}_{j-q})$$

$$+ \square_{jq}(\phi^*_{jq}\phi_{jq} - \phi^*_{j-q}\phi_{j-q})] - \frac{1}{8}\, 3b_1\phi_1(\phi^*_{1q}\phi^*_{1-q} + \phi_{1q}\phi_{1-q})$$

$$+ \Lambda_{12}(\phi^*_{1q}\phi^*_{2-q} + \phi_{1q}\phi_{2-q}) + \tilde{\Lambda}_{12}(\phi^*_{1q}\phi_{2q} + \phi^*_{2q}\phi_{1q})$$

$$+ \Lambda_{23}(\phi^*_{2q}\phi^*_{3-q} + \phi_{2q}\phi_{3-q}) + \tilde{\Lambda}_{23}(\phi^*_{2q}\phi_{3q} + \phi^*_{3q}\phi_{2q})$$

$$+ \Lambda_{31}(\phi^*_{3q}\phi^*_{1-q} + \phi_{3q}\phi_{1-q}) + \tilde{\Lambda}_{31}(\phi^*_{3q}\phi_{1q} + \phi^*_{1q}\phi_{3q})\}\,, \qquad (3)$$

where V_0 corresponds to the static equilibrium state, that is, when we substitute $\phi_j(\vec{r}) = \phi_j$ $(j = 1,3)$ into (2). The coefficients of (3) are abbreviated as follows:

$$\Lambda_{11} = e_0[(\vec{Q}_1 . \vec{q})^2 + Q_1^2(\frac{1}{3}K_1 - Q_1)] + f_0(\vec{Q}_1 \times \vec{q})^2 + (\frac{1}{2}\, 3c_0 + d_0)(\phi_2^2 + \phi_3^2) \qquad (4)$$

$$\Lambda_{22} = e_0\{(\vec{Q}_2 . \vec{q})^2 + [\vec{Q}_2 . (\frac{1}{3}\vec{K}_2 - \vec{k} - \vec{Q}_2)]^2\} + f_0[(\vec{Q}_2 \times \vec{q})^2 + (\vec{Q}_2 \times \vec{k})^2]$$
$$+ (\frac{1}{2}\, 3c_0 + d_0)(\phi_3^2 + \phi_1^2) \qquad (5)$$

$$\Lambda_{33} = e_0\{(\vec{Q}_3 . \vec{q})^2 + [\vec{Q}_3 \cdot (\frac{1}{3}\vec{K}_3 - \vec{k} - \vec{Q}_3)^2]\} + f_0[(\vec{Q}_3 \times \vec{q})^2 + (\vec{Q}_3 \times \vec{k})^2]$$
$$+ (\frac{1}{2}\, 3c_0 + d_0)(\phi_1^2 + \phi_2^2) \qquad (6)$$

$$\tilde{\Lambda}_{12} = (\frac{1}{2}\, 3c_0 + d_0)\phi_1\phi_2 \qquad (7)$$

$$\Lambda_{12} = \tilde{\Lambda}_{12} - \frac{1}{4}\, 3b_0\phi_3 \qquad (8)$$

$$\tilde{\Lambda}_{23} = (\frac{1}{2}\, 3c_0 + d_0)\phi_2\phi_3 \qquad (9)$$

$$\Lambda_{23} = \tilde{\Lambda}_{23} - \frac{1}{4}\, 3b_0\,\phi_1 \qquad (10)$$

$$\tilde{\Lambda}_{31} = (\frac{1}{2}\, 3c_0 + d_0)\, \phi_3\phi_1 \tag{11}$$

$$\Lambda_{31} = \tilde{\Lambda}_{31} - \frac{1}{4}\, 3b_0\, \phi_2 \tag{12}$$

$$\square_{11} = e_0\vec{Q}_1 . (\frac{1}{3}\, \vec{k}_1 - \vec{Q}_1)\, (\vec{Q}_1 . \vec{q}) \tag{13}$$

$$\square_{22} = e_0\, [\vec{Q}_2 . (\frac{1}{3}\, \vec{k}_2 - \vec{k} - \vec{Q}_2)\, (\vec{Q}_2 . \vec{q})] - f_0\, (\vec{Q}_2\times\vec{k})\, (\vec{Q}_2\times\vec{q}) \tag{14}$$

$$\square_{33} = e_0\, [Q_3 . (\frac{1}{3}\, \vec{k}_3 + \vec{k} - \vec{Q}_3)\, (\vec{Q}_3 . \vec{q})] + f_0\, (\vec{Q}_3\times\vec{k})\, (\vec{Q}_3\times\vec{q}) \tag{15}$$

We consider the particular case when $\vec{q}$ is perpendicular to $\vec{Q}$, and compute the eigenmode frequencies using the symmetric and anti-symmetric combinations of coupled modes[1,2]. In these calculations we have used some symmetry simplifications and the minimization conditions in terms of k, ϕ_1 and ϕ_2, for the stripe geometry. These lead to a vanishing of the phase-amplitude cross term of (3), which in this case can be written as

$$V = V_0 + V^+ + V^- \quad , \tag{16}$$

where V_0 has already been defined,

$$\begin{aligned} V^+ = \frac{1}{2} \sum_q \{ \sum_j [\frac{1}{2}\, 3c_0\phi_j^2 + e_0\, (\vec{Q}_j . \vec{q})^2 + f_0\, (\vec{Q}_j\times\vec{q})^2] A^*_{jq} A_{jq} \\ + \frac{1}{4}\, 3[(b_0\phi_1^{-1}\phi_2^2 - \frac{1}{2}\, b_1\phi_1) A^*_{1q} A_{1q} \\ + b_0\phi_1 \sum_{j=2}^{3} A^*_{jq} A_{jq}] + [(3c_0+2d_0)\phi_1\phi_2 - \frac{1}{4}\, 3b_0\phi_2]\, (A^*_{1q} A_{2q} + A^*_{2q} A_{1q} \\ + A^*_{3q} A_{1q} + A^*_{1q} A_{3q}) + [(3c_0+2d_0)\phi_2^2 - \frac{1}{4}\, 3b_0\phi_1]\, (A^*_{2q} A_{3q} + A^*_{3q} A_{2q}) \} \end{aligned} \tag{17}$$

and

$$\begin{aligned} V^- = \frac{1}{2} \sum_q i \sum_j [e_0\, (\vec{Q}_j . \vec{q})^2 + f_0\, (\vec{Q}_j\times\vec{q})^2] P^*_{jq} P_{jq} \\ + \frac{1}{4}\, 3[(b_0\phi_1^{-1}\phi_2^2 + \frac{1}{2}\, 3b_1\phi_1) P^*_{1q} P_{1q} + b_0\phi_1 \sum_{j=2}^{3} P^*_{jq} P_{jq}] \\ + \frac{1}{4}\, 3b_0\phi_2\, (P^*_{1q}P_{2q} + P^*_{2q}P_{1q} + P^*_{3q}P_{1q} + P^*_{1q}P_{3q}) + \frac{1}{4}\, 3b_0\phi_1\, (P^*_{2q}P_{3q} + P_{3q}P^*_{2q}) \} . \end{aligned} \tag{18}$$

We diagonalize the two matrices obtained from (17) and (18) and find six eigenmode: five optical (three amplitudons with A_1, A_1 and B_1 symmetries, respectively, and two phasons with B_1 symmetry. The results for the B_1 modes are, in the case of amplitudons:

$$B_1 : \frac{1}{2}\, 3b_0\phi_1 - (\frac{1}{2}\, 3c_0 + 2d_0)\phi_2^2 + \frac{1}{4}\,(3e_0 + f_0)Q_2^2 q^2 \;; \tag{19}$$

and in the case of phasons:

$$B_1 \;: \frac{1}{4}\,(3e_0+f_0)Q_2^2 q^2 \;. \tag{20}$$

The algebraic manipulations involving the A_1 modes are more complicated but it is possible to obtain them exactly to order q^2, in terms of different parameters. For the case of amplitudons (A_1, A_1), we find

$$A_1 \;:\; \frac{1}{2}\;\{\alpha+\beta+\gamma - \left[(\alpha+\beta\pm\gamma)^2 + 8\nu^2\right]^{1/2}\} \tag{21}$$

and for the case of phasons (A_1, A_1), we have

$$A_1 \;:\; \frac{1}{2}\{\tilde{\alpha}+\tilde{\beta}+\tilde{\gamma} - \left[(\tilde{\alpha}+\tilde{\beta}\pm\tilde{\gamma})^2 + 8\tilde{\nu}^2\right]^{1/2}\} \;. \tag{22}$$

In (21) and (22) we use the following parameters:

$$\alpha = \frac{1}{2}\, 3c_0\phi_0^2 + \tilde{\alpha} \;;\quad \tilde{\alpha} = \frac{1}{4}\, 3b_0\phi_1 + \frac{1}{4}(3e_0+f_0)Q_2^2 q^2 \;;\quad \beta = (3c_0+2d_0)\phi_2^2 - \tilde{\beta} \;;$$

$$\tilde{\beta} = \frac{1}{4}\, 3b_0\phi_1 \;;\quad \nu = (3c_0+2d_0)\phi_1\phi_2 - \tilde{\nu} \;;\quad \tilde{\nu} = \frac{1}{4}\, 3b_0\phi_2 \;;$$

$$\gamma = \frac{1}{4}\, 3b_0\phi_1^{-1}\phi_2^2 - \frac{1}{8}\, 3b_1\phi_1 + \frac{1}{2}\, 3c_0\phi_1^2 + f_0 Q_1^2 q^2 \;;$$

$$\tilde{\gamma} = \frac{1}{4}\, 3b_0\phi_1^{-1}\phi_2^2 + \frac{1}{8}\, 9b_0\phi_1 + f_0 Q_1^2 q^2 \;. \tag{23}$$

Similarly[1], A_1 and B_1 are the symmetry characters of the irreducible representations of the group C_{2V}-2mm and expressions (19) - (22), represent $M^*\omega^2/4$, where ω and M^*, are defined in ref. 1. Table 1 shows the correlation between the symmetry characters of space group D_{6h}-6/mmm and C_{2V} - 2mm.

Another possibility is to take the direction of $\vec{q}$ parallel to $\vec{Q}$, where there is a q-linear amplitude-phase cross term in (16), that is, we have an additional term $\overset{\pm}{V}$, which we will not discuss here.

Table 1 - Correlation between irreducible representations of the symmetry groups of amplitudon (A) and phason (P) modes of the hexagonal and orthorhombic stripe incommensurate phases of $2H$-$TaSe_2$.

	Hexagonal (D_{6h}-6/mmm)		Stripe (C_{2V} - 2mm)
(A)	E_{2g}	→	A_1
		→	B_1
	A_{1g}	→	A_1
(B)	E_{1u}	→	A_1
		→	B_1
	B_{1u}	→	A_1

3. CONCLUSIONS

In spite of considerable efforts in the last years, more theoretical work must be done in order to understand the fascinating variety of CDW phases in $2H$-$TaSe_2$. Our calculations of the normal mode frequencies of OSICDW for this compound give a good contribution in this direction. However the failure in observing some of the normal modes has prevented the determination of some important parameters in the theory[7], Because of these problems, we cannot very the theoretical results against experimental data we quote in passing that analytical computations have been done, using a procedure similar to that of the precedent paper[1]. In this case, the algebraic calculation is not straightforward. We have derived the equilibrium conditions and finally obtained the eigenmode frequencies for both amplitudons (A_1, B_1) and phasons (A_1, B_1) respectively. We have only considered one possibility for the $\vec{q}$-direction. Finally we must emphasise that new experimental information about that this phase will be necessary in order to verify the theoretical predictions.

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Resumo

Cálculos analíticos são desenvolvidos para obter os modos normais da fase densidade de onda de carga incomensuravel "stripe" ortorrômbica (OSICDW) do metal de transição $2H\text{-}TaSe_2$, usando a teoria McMillan-Landau de transição de fase. Os amplitudons e phasons são obtidos numa particular direção, onde é possível conseguir-se expressões mais simples para os mesmos.