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ICDW Eigenmode Frequencies of $2H\text{-}TaSe_2$ in McMillan-Landau Theory

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Abstract Analytical and numerical calculations are performed in orderto obtain order-parameter fluctuation modes for the triple hexagonal incommensurate charge-density-wave (ICDW) phase in $2H\text{-}TaSe_2$, using the McMillan-Landau Theory of Phase transitions. It is shown that the optical modes present large energy gaps, except when T is very close to onset temperature, T_0.

1. INTRODUCTION

Very recently many theoretical and experimental works[1-4] have concentrated on the study of the commensurate phase of the dichalcogenide transition metal $2H\text{-}TaSe_2$, owing to the controversy whether such phase is orthorhombic rather than hexagonal. Other authors have pointed out the possibility in considering the incommensurate phase present in this substance, between $112^{o}K$ and $122^{o}K$ as being a typical honeycomb (hexagonal) phase. Despite this, Chen et *al*[5] have emphasized that we can experimentally observe that in this range the hexagonal symmetry is retained on a larger scale.

In this note we report some calculations of fluctuation eigenmodes (amplitudons and phasons) for the hexagonal incommensurate phase of $2H\text{-}TaSe_2$, using the McMillan-Landau free energy, which is very convenient in the study of incomrnensurate phases[6]. Firstly, we summarize the basic points of McMillan's approach, than we write the free energy for this phase and finally we compute the fluctuation eigenmode.

2. THEORY

In 1975, McMillan introduced his Landau free energy potential,

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where the real order parameter $\alpha(\vec{r})$ is obtained from the electronic charge density $\rho(\vec{r})$. After some algebraic manipulations it is possible to expand $\alpha(\vec{r})$ in terms of three complex order parameters $\psi_j(\vec{r})$, with amplitudes $\phi_j(\vec{r})$, which correspond to each of the three CDW's present in $2H$-$TaSe_2$.

In this theory one normally defines the following vectors: $\vec{k}_j$ $(j = 1,6)$ are the six shortest reciprocal lattice vectors in the hexagonal symmetry; $\vec{Q}_j$ $(j = 1,3)$ are the three incommensurate vectors. These three vectors lie in directions 120^o apart. The other vector $\vec{q}_j$ $(j = 1,3)$ represent the charge-density-wave vectors and q are the excitation wave vectors,

Now, we consider the hexagonal incommensurate phase of $2H$-$TaSe_2$. This structure is characterized by the space group D_{6h}-6/mmm. We perform the calculations of excitation modes, which have been previously considered by McMillan[7]. That author has done computations of order-parameter fluctuation modes for this phase, discussing only the case of phase fluctuation modes (phasons) with wave-vector $\vec{q}$ along a symmetry axis. In our case, we concentrate on amplitude fluctuation modes (amplitudons) for a different $\vec{q}$-direction, doing the same for the case of phasons. In our discussion we have confined ourselves only to intralayer terms.

In this phase, the McMillan-Landau free energy can be rewritten as

$$V = \frac{1}{2}\int d^2r\{\sum_j \left[a_0|\phi_j|^2 + \frac{1}{4}\,3c_0|\phi_j|^4 + 2e_0|\vec{Q}_j\cdot\vec{\nabla}\phi_j|^2 + 2f_0|\vec{Q}_j\times\vec{\nabla}\phi_j|^2\right]$$
$$- \frac{1}{2}\,3b_0(\phi_1\phi_2\phi_3+\phi_1^*\phi_2^*\phi_3^*) + (3c_0 + 2d_0)(|\phi_1\phi_2|^2+|\phi_2\phi_3|^2+|\phi_3\phi_1|^2)\} \tag{1}$$

because, $\vec{q}_j = \vec{Q}_j$, and $\phi_1 = \phi_2 = \phi_3 = \phi_0$. The vectors being incommensurate with the lattice, only the uniform terms[7] a_0, b_0, c_0, ..., contribute. The 'lock-in' cubic term vanishes in this case. The first term in eq.(1) is the unscreened elastic constant; the second is the quartic energy of Landau expansion. The third and fourth terms are the elastic energy contributions; the fifth one is a weak CDW interaction permitted by symmetry and the last term is a CDW interaction which arise when two CDW's compete to open an energy gap on the same portion of Fermi surface[7].

Now, we use the minimum value for the free energy potential in terms of ϕ_0, in order to expand eq. (1) in powers of ϕ_{jq}, keeping only second-order terms,

$$V = V(\phi_0) + \sum_{jq} \frac{1}{2} \left[a_0 + 9c_0\phi_0^2 + 4d_0\phi_0^2 + 2e_0(\vec{Q}.\vec{q})^2 + 2f_0(\vec{Q}\times\vec{q})^2\right]\phi^*_{jq}\phi_{jq}$$

$$+ \frac{1}{8} 3c_0\phi_0^2 \sum_{jq} (\phi^*_{jq}\phi^*_{j-q} + \phi_{jq}\phi_{j-q}) - \frac{1}{2}\left[\frac{1}{2} 3b_0\phi_0\right.$$

$$\left. - (3c_0 + 2d_0)\phi_0^2\right] \sum_q (\phi^*_{1q}\phi^*_{2-q} + \phi_{1q}\phi_{2-q} + \text{permutations})$$

$$+ \frac{1}{2} (3c_0 + 2d_0)\phi_0^2 \sum_q (\phi^*_{1q}\phi_{2q} + \phi^*_{2q}\phi_{1q} + \text{permutations}) \tag{2}$$

This expression is a little different from one in McMillan[8], because he omitted the contribution of the last term of eq.(1). We will retain it here, because it is important in the case of amplitude fluctuation mode calculations.

Following the standard procedure, we compute the eigenmodes, rewritting eq, (2) in terms of the coupled modes A_{jq} and P_{jq}, such that

$$V = V(\phi_0) + V^+ + V^- \tag{3}$$

where V factorizes into amplitude and phase contributions with no cross term and $V(\phi_0)$ corresponds to the minimum of the free energy. So,

$$V^+ = \frac{1}{2}\{\sum_{jq} \left[\frac{1}{4} 3b_0\phi_0 + \frac{1}{2} 3c_0\phi_0^2 + e_0(\vec{Q}_j.\vec{q})^2 + f_0(\vec{Q}_j\times\vec{q})^2\right]A^*_{jq}A_{jq}$$

$$-\left[\frac{1}{4} 3b_0\phi_0 - (3c_0+2d_0)\phi_0^2\right] \sum_q (A^*_{1q}A_{2q} + A^*_{2q}A_{1q} + \text{permutations}) \tag{4}$$

$$V^- = \frac{1}{2}\{\sum_{jq} \left[\frac{1}{4} 3b_0\phi_0 + e_0(\vec{Q}_j.\vec{q})^2 + f_0(\vec{Q}_j\times\vec{q})^2\right]P^*_{jq}P_{jq}$$

$$+ \frac{1}{4} 3b_0\phi_0 \sum_q (P^*_{1q}P_{2q} + P^*_{2q}P_{1q} + \text{permutations}) \tag{5}$$

Now we diagonalize the matrices obtained from eqs. (4) and (5) find the six mode frequencies, four being optical, which are three amplitudons and one phason; two are acoustic modes, which are both phasons.

We have considered a particular $\vec{q}$ direction, when this vector is perpendicular to $\vec{Q}_1$, writing results correct to order q^2. In this case, we find the amplitudons,

$$E_{2g} : \frac{1}{2} 3b_0\phi_0 - (\frac{1}{2} 3c_0 + 2d_0)\phi_0^2 + \frac{1}{4} (3e_0 + f_0)Q_1^2q^2 \qquad (6)$$

$$E_{2g} : \frac{1}{2} 3b_0\phi_0 - (\frac{1}{2} 3c_0 + 2d_0)\phi_0^2 + \frac{1}{4} (e_0 + 3f_0)Q_1^2q^2 \qquad (7)$$

$$A_{2g} : - \frac{1}{4} 3b_0\phi_0 + (\frac{1}{2} 15c_0 + 4d_0)\phi_0^2 + \frac{1}{2} (e_0+f_0)Q_1^2q^2 \qquad (8)$$

Similarly, we can compute the phasons and find

$$E_{1u} : \frac{1}{4} (3e_0 + f_0)Q_1^2q^2 \qquad (9)$$

$$E_{1u} : \frac{1}{4} (e_0 + 3f_0)Q_1^2q^2 \qquad (10)$$

$$B_{1u} : \frac{1}{4} 9b_0\phi_0 + \frac{1}{2} (e_0+f_0)Q_1^2q^2 \qquad (11)$$

E_{1u}, A_{1u}, E_{2g} and A_{1g} are the symmetry character of the irreducible representation of the D_{6h}-6/mmm grcup and each expression above represents $M^*\omega^2/4$, where ω is the mode frequency[7]. These results (6-11) are illustrated numerically in figures 1 and 2. The value of M^* = 206 au [7].

3. NUMERICAL RESULTS

In this section we present the results of numerical computations of normal mode frequencies (amplitudons and phasons) for the hexagonal incommensurate superlattice of a single layer of the $2H-TaSe_2$ structure. We have calculated the mode frequencies in terms of the wave -vector $\vec{q}$ and we have also obtained the behaviour of these modes as the temperature changes, Firstly, we have followed Nakanish and Shiba[9], using their values for the phenomenological parameters b_0, b_1 and c_0 in order to simulate 2H-$TaSe_2$. Those authors have taken b_0 = 4/30, b_1 = 1.2 and c_0 = 8/3. However, as regards d_0, we have taken a different value[6], namely d_0 = -3.9, corresponding to our use of a different form of free energy potential (1). We have performed straightforward analytical manipulations, obtained the eigenmode frequencies, that is, amplitudons (E_{2g}, A_{1g}) and phasons (E_{1u}, B_{1u}) in the range of temperature between 122°K and 112°K, where experimentally the charge-density-wave phase has hexagonal symmetry. These parameter values have been used with success in fitting the experimental data[10] involving other features of diifferent phases of this compound[6].

Figures (1) and (2) show the eigenmode frequencies from the onset temperature T_0 (122°K), where the Landau parameter a_0 = 0, until the hexagonal-orthorhombic stripe incommensurate transition temperature (112°K) where, in this numerical simulation[6], a_0 = 0.5499. These figures are based on (6-11) with $f_0 = 0$ and the coordinates have been expressed as $= \frac{1}{2} M^{*1/2}\omega$ and $\tilde{q} = e^{1/2} Q_1 q$.

4. CONCLUSION

The theoretical description of the excitations of the lattice below a phase transformation from a normal structure to an incommensurate one is not straightforward, because translational symmetry is lost. Despite this aspect, it is very important to increase efforts in order to get information about incomnensurate rnaterials like 2H-$TaSe_2$. Our numerical investigation has concentrated in simulating this compound[6], assuming phenomenological parameters to obtain some picture of normal-mode frequencies of the hexagonal incommensurate phase as the temperature changes. The parameter values used in this work are in good agreement with experimental data[10]. In the case of this phase, the 'lock-in' (Umklapp) cubic term, in the McMillan-Landau free energy, drops out. The basic points about the phason modes in this phase are that there are two hydrodynamic modes and another optical one with an energy gap (1/4) $9b_0\phi_0$. In our calculations we have considered one simple possibility for the $\vec{q}$-direction. These modes involve long-wavelength distortions of the charge-density lattice, Another aspect is that ths more

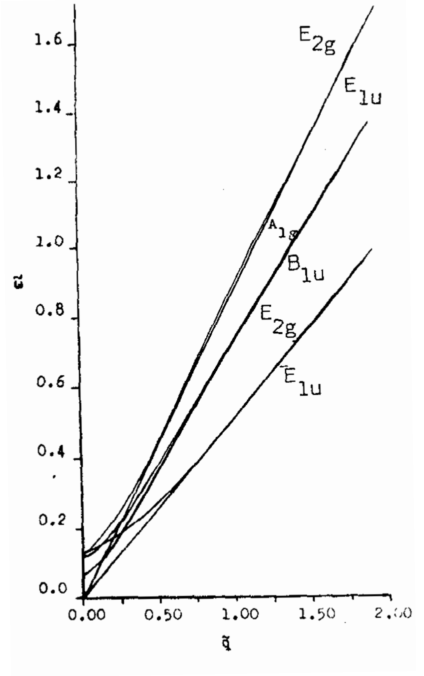

Fig. 1 - Normal modes of the hexagonal incommensurate CDW, with $\omega = 0.5\ M^{1/2}\omega$, $\tilde{q} = e_0^{1/2}\ Q_1 q$ and T = 122°K.

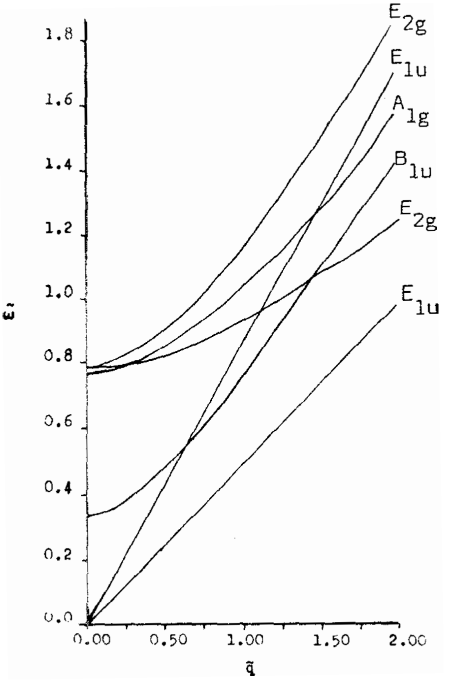

Fig.2 - Normal modes of thehexagonal incommensurate CDW, with T = 112°K. Coordinate axes are the same as in fig.1.

accessible characteristics of the pictures involving the amplitudonsand phasons, are the variations of the energy gaps for the optic modes (A_{1g}, E_{2g}, B_{1u}).

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Resumo

Cálculos analíticos e numéricos são efetuados com o fito de obter modos de flutuação do parâmetro de ordem para a fase densidade de onda de carga incomensuravel hexagonal tripla no $2H\text{-}TaSe_2$, usando a Teoria McMillan-Landau de transição de fase. É mostrado que os modos Ópticos apresentam largos "Gaps" de energia, excetuando-se quandoa temperatura é muito próxima daquela entre a fase normal e incomensuravel T_0.