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New Calculations of ICDW Eigenmode Frequencies of 2H-$TaSe_2$

A. Ribeiro Filho, J. F. M. Rocha, E. M. do Nascimento, and D. S. de Vasconcelos

Instituto de Física, Universidade Federal da Bahia, Campus Universitário de Ondina, 40210-340, Salvador, BA, Brazil

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Using an extended expression for the Landau-Lifshitz free-energy density we have computed order-parameter fluctuation eigenmodes for the triple hexagonal incommensurate charge-density-wave (ICDW) phase in 2H-$TaSe_2$. Phasons and amplitudons have been obtained for two particular directions and we have performed some numerical calculations.

1. Introduction

Many theoretical and experimental works have emphasized the importance of the phenomenon of charge-density-waves (CDWs) in solids, which has received a great deal of attention from condensed matter physics researchers in recent years. A large quantity of materials with a highly anisotropic band structure develops CDW. Organic conductors (tetrathiafulvalene–tetracyanoquinodimethane: TTF–TCNQ), inorganic layer compounds, various semiorganic compounds (like postassium platinocyanide: KCP), as well as inorganic linear-chain compounds ($NbSe_3$, $TaSe_3$, ...) are good examples of such systems [1 to 5]. Grüner [2] has stressed that the formation of CDW ground states is by now well discussed in a broad range of so-called low-dimensional solids.

Very recently, many authors have discussed distinct problems involving CDW. Latyshev et al. [4] studying Aharonov-Bohm effect (ABE) on CDW in $NbSe_3$, have observed oscillations of the CDW conduction as a function of magnetic field due to the CDW passing columnar defects containing trapped magetic flux. These authors have stressed that such observation is consistent with the instanton ABE predicted in CDW systems. In a recent work, Montambaux [5] has shown that, in an appropriate geometry, the dephasing of the electron–hole pair may lead to the suppression of the CDW or spin-density-wave state by an applied electric field.

Using a sophisticated Landau-Lifshitz free-energy density, in order to discuss different properties of 2H-$TaSe_2$, McMillan [6] was successful in introducing the notion of discommensuration defect or soliton-like behaviour of the incommensurate CDW near the incommensurate–commensurate "lock-in" phase transition despite the existence of alternative theoretical models. Wilson and Yoffe [3] have pointed out that the transition metal 2H-$TaSe_2$ has a layered structure, in each layer a hexagonal sheet of Ta ions is sandwiched between two hexagonal sheets of Se ions.

The order parameter fluctuation modes (phasons and amplitudons) for the CDW in incommensurate systems have been studied by other authors and, in the case of 2H-$TaSe_2$, there has been a great deal of work due to the complexity of its phase transition picture [7]. The experimental work performed by Steinitz and Genossar [8] improved substantially the understanding of the incommensurate superlattice of this substance, inferring that in the range between 112 and 90 K, this superlattice presents two different phases. The first with hexagonal symmetry exhibits at 112 K a first-order phase transition to an orthorombic stripe phase, which subsequently "locks-in" at 90 K. Despite the controversy around the commensurate (hexagonal or orthorhombic symmetry) phase, meanwhile between 112 and 122 K, this substance presents an incommensurate phase characterized by the space group $D_{6h}-6/mmm$, that is, in this range the hexagonal symmetry is retained on a larger scale [7,8].

In this work we report some calculations of amplitudons and phasons using an extended expression for the Landau-Lifshitz free energy density keeping terms up to sixth order [9]. In Section 2 we present calculations involving the ICDW phase, maintaining the same notation of Rocha et al. [9]. In Section 3 we will perform calculations involving eigenmodes frequencies and providing new analytical results, for two distinct directions of the excitation wave vector ($\mathbf{q}$), for the case of phasons and amplitudons, respectively. In Section 4 we present some numerical results, which are illustrated by different figures and, finally, in Section 5 we discuss the results of this work.

2. Theoretical Formalism

In order to perform the eigenmode frequencies calculations, we write the extended expression for the Landau-Lifshitz free-energy density

$$V = \int \mathrm{d}^2 r \left[\sum_{j=2}^{6} (-1)^j \tilde{a}_j \alpha^j + \tilde{a}_7 \sum_{j=1}^{3} |(\mathbf{Q}_j - \nabla - iQ_j^2)\, \psi_j|^2 \right. \\ \left. + \tilde{a}_8 \sum_{j=1}^{3} |\mathbf{Q}_j \times \nabla \psi_j|^2 + \tilde{a}_9 (|\psi_1 \psi_2|^2 + |\psi_2 \psi_3|^2 + |\psi_3 \psi_1|^2) \right], \tag{1}$$

where $\tilde{a}_j$ $(j = 2, \ldots, 9)$ are the phenomenological parameters, with $\tilde{a}_5 = 0$. The real order parameter $\alpha(\mathbf{r}) = \mathrm{Re} \sum_{j=1}^{3} \psi_j(\mathbf{r})$ is obtained from the electronic charge density $\varrho(\mathbf{r})$, and $\psi_j(\mathbf{r})$ are complex order parameters linked to the three components of the triple charge-density-waves occurring in this material [9]. Normally, we define the following vectors: $\mathbf{Q}_j$ $(j = 1, 2, 3)$ are the three incommensurate vectors, which lie in directions 120° apart; $\mathbf{G}_j$ $(j = 1, \ldots, 6)$ are the six shortest reciprocal lattice vectors in the hexagonal symmetry; $\mathbf{q}_j$ $(j = 1, 2, 3)$ are the charge-density-wave vectors and $\mathbf{q}$ are the excitation wave vectors. We must remember from [6, 9] that

$$\tilde{a}_j(\mathbf{r}) = \tilde{a}_{0j}(\mathbf{r}) + \tilde{a}_{1j}(\mathbf{r}) \sum_{j=1}^{3} \exp(i\mathbf{G}_j \cdot \mathbf{r}). \tag{2}$$

Let us consider the hexagonal incommensurate phase of $2H\text{-}TaSe_2$. We have performed the calculations of excitation modes, which have been previously considered by McMillan [6], despite that author has only discussed phasons (phase fluctuation modes) with wave-vector $\mathbf{q}$ along a symmetry axis. In this work, we report our calculations on amplitude fluctuation modes (amplitudons) for two different $\mathbf{q}$-directions, doing the same for the case of phasons, and considering a new expression for the free-energy density.

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

$$V = \frac{1}{2} \int \mathrm{d}^2 r \Bigg\{ \sum_{j=1}^{3} [\tilde{a}_{02} \, |\psi_{0j}(\mathbf{r})|^2 + \frac{1}{4} \, 3\tilde{a}_{04} \, |\psi_{0j}(\mathbf{r})|^4 + \frac{1}{8} \, 5\tilde{a}_{06} \, |\psi_{0j}(\mathbf{r})|^6$$
$$+ 2\tilde{a}_{07} \, |(\mathbf{Q}_j \cdot \nabla \psi_{0j}(\mathbf{r})|^2 + 2\tilde{a}_{08} \, | \, \mathbf{Q}_j \times \nabla \psi_{0j}(\mathbf{r})|^2] - \frac{1}{2} \, 3\tilde{a}_{03} [\psi_{01}(\mathbf{r}) \, \psi_{02}(\mathbf{r}) \, \psi_{03}(\mathbf{r})$$
$$+ \psi^*_{01}(\mathbf{r}) \, \psi^*_{02}(\mathbf{r}) \, \psi^*_{03}(\mathbf{r})] + (3\tilde{a}_{04} + 2\tilde{a}_{09}) \, [|\psi_{01}(\mathbf{r}) \, \psi_{02}(\mathbf{r})|^2 + |\psi_{02}(\mathbf{r}) \, \psi_{03}(\mathbf{r})|^2$$
$$+ |\psi_{03}(\mathbf{r}) \, \psi_{01}(\mathbf{r})|^2] + \frac{1}{2} \, 45\tilde{a}_{06} (|\psi_{01}(\mathbf{r}) \, \psi_{02}(\mathbf{r}) \, \psi_{03}(\mathbf{r})|^2 + \frac{1}{4} [|\psi_{01}(\mathbf{r})|^4 \, (|\psi_{02}(\mathbf{r}))|^2$$
$$+ |\psi_{03}(\mathbf{r})|^2) + |\psi_{02}(\mathbf{r})|^4 \, (|\psi_{03}(\mathbf{r})|^2 + |\psi_{01}(\mathbf{r})|^2) + |\psi_{03}(\mathbf{r})^4 \, (|\psi_{01}(\mathbf{r})|^2 + |\psi_{02}(\mathbf{r})|^2)]) \Bigg\} \, . \quad (3)$$

We can observe that the main contributions for the free energy in Eq. (3) are of the uniform terms from expresssion (2), that is, $\tilde{a}_{02}$, $\tilde{a}_{03}$, $\tilde{a}_{04}, \ldots$ In this case the cubic "lock-in" term vanishes. The first three terms in the equation above are typical of the Landau free-energy expansion. The fourth and fifth terms are contributions from the elastic energy, while the sixth and seventh ones indicate the interaction between the charge-density-waves. The other terms are also resulting from interactions between CDWs and they have arosen in this equation due to the inclusion of the sixth order term in the homogeneous part of the free-energy potential expression. We have also seen that in the Eq. (3) two types of quartic terms arise, and that if $|\tilde{a}_{09}| = \frac{1}{2} 3\tilde{a}_{04}$ ($\tilde{a}_{09} < 0$), the first quartic term vanishes and there is no quartic order interaction energy between the three CDWs.

In order to make simple our calculations we have considered, like other authors [6,10], the simple case where the CDW amplitudes are equal and constant, that is $\psi_{01}(\mathbf{r}) = \psi_{02}(\mathbf{r}) = \psi_{03}(\mathbf{r}) = \psi_0$. In this case, we can integrate Eq. (3) on unit area, such that

$$V(\psi_0) = \frac{1}{2} \, 3 \big[\tilde{a}_{02} - \tilde{a}_{03}\psi_0 + \frac{1}{4} \big(15\tilde{a}_{04} + 8\tilde{a}_{09}\big) \, \psi_0^2 + \frac{1}{8} 155\tilde{a}_{06}\psi_0^4\big] \, \psi_0^2 \, . \quad (4)$$

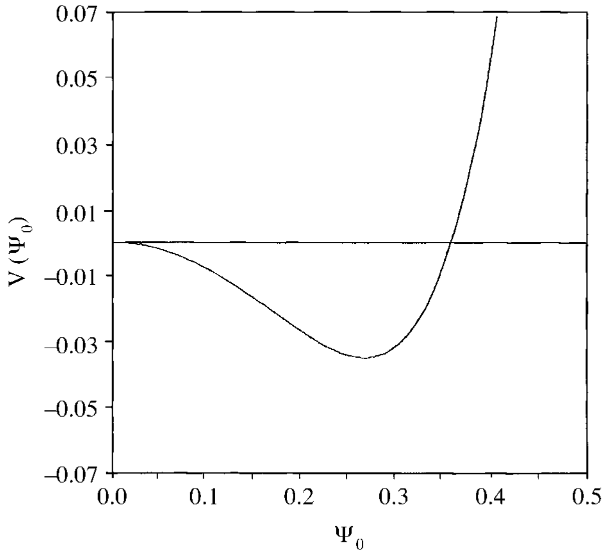

Fig. 1. Incommensurate free energy $V(\psi_0)$ versus order parameter ψ_0, with $\tilde{a}_{06} = 1$, $T_0 = 112\,\mathrm{K}$. The other phenomenological parameters are given in the text

Fig. 1 shows the generalized free energy, which reproduces McMillan's results [6] when $\tilde{a}_{06} = 0$. In Eq. (4) we can see the cubic "lock-in" term where we consider $\tilde{a}_{03} > 0$, because this term should contribute for the free-energy reduction. The extremals of the potential (4) are given by $\psi_0 = 0$, and by solutions of the following equation:

$$2\tilde{a}_{02} - 3\tilde{a}_{03}\psi_0 + (15\tilde{a}_{04} + 8\tilde{a}_{09})\,\psi_0^2 + \tfrac{1}{4}\,465\tilde{a}_{06}\psi_0^4 = 0\,, \tag{5}$$

which gives the other order parameter values that minimize the free energy and compute the transition temperature between the normal and incommensurate phases in 2H-$TaSe_2$.

3. Amplitudons and Phasons Calculations

In this section we compute the order parameter fluctuation modes for the triple hexagonal incommensurate charge-density-wave in 2H-$TaSe_2$. We will assume the phase and amplitude of the order parameter to vary, and we use the Lagrangian mechanics background in order to consider the oscillations associated to the triple CDWs. So, the free energy given in Eq. (4) is considered as the potential energy of the system, and the kinetic energy is expressed by $T = \int \mathrm{d}^2 r\, \frac{1}{2} \sum_{i,j=1}^{3} T_{ij} (\partial\psi_{0i}/\partial t)\,(\partial\psi_{0j}^*/\partial t)$, where (T_{ij}) is a symmetric matrix. The Lagrangian is given by $L = T - V$, and the components of our order parameter have been considered as components of a classical field and can be rewritten as

$$\psi_{0j}(\mathbf{r},\, t) = \psi_{0j} + \eta_j(\mathbf{r},\, t)\,, \tag{6}$$

where $\eta_j(\mathbf{r},\, t)$ is a small deviation of the amplitude, $\psi_{0j} = \psi_0$ $(j = 1, 2, 3)$, of the static CDW. However, η_j are periodic, such that we can expand them in Fourier series, that is, $\eta_j = \sum_q \exp{(i\mathbf{q}\cdot\mathbf{r})}\,\psi_{0jq}(t)$, finding directly the Euler-Lagrange equation for the Fourier amplitudes $\psi_{0jq}(t)$ or, equivalently, to work with the quadratic Lagrangian in $\psi_{0jq}(\mathbf{r},\, t)$ from beginning, so we can substitute

$$\psi_{0j}(\mathbf{r},\, t) = \psi_0 + \sum_q \exp{(i\mathbf{q}\cdot\mathbf{r})}\,\psi_{0jq}(t) \tag{7}$$

in the Lagrangian L. After integrating we find the following expressions for the kinetic and potential energies, respectively:

$$T = \sum_q \sum_{i,j=1}^{3} \tfrac{1}{2} T_{ij}\, \dot{\psi}_{0iq}^*(t)\, \dot{\psi}_{0jq}(t) \tag{8}$$

and

$$\begin{aligned} V = V(\psi_0) &+ \sum_q \sum_{j=1}^{3} \tfrac{1}{2} \big[\tilde{a}_{02} + (9\tilde{a}_{04} + 4\tilde{a}_{09})\,\psi_0^2 + \tfrac{1}{8}\,675\tilde{a}_{06}\psi_0^4 \\ &+ 2\tilde{a}_{07}(\mathbf{Q}_j\cdot\mathbf{q})^2 + 2\tilde{a}_{08}(\mathbf{Q}_j\times\mathbf{q})^2\big]\,\psi_{0jq}^*\psi_{0jq} \\ &+ \tfrac{1}{8}\,3\big(\tilde{a}_{04} + \tfrac{1}{2}\,105\tilde{a}_{06}\psi_0^2\big)\,\psi_0^2 \sum_{jq} \big(\psi_{0jq}^*\psi_{0j-q}^* + \psi_{0jq}\psi_{0j-q}\big) \\ &- \tfrac{1}{2}\big[\tfrac{1}{2}\,3\tilde{a}_{03}\psi_0 - (3\tilde{a}_{04} + 2\tilde{a}_{09} - 45\tilde{a}_{06})\,\psi_0^2\big] \sum_q (\psi_{01q}^*\psi_{02-q}^* + \psi_{01q}\psi_{02-q} + \text{permutations}) \\ &+ \tfrac{1}{2}\big[(3\tilde{a}_{04} + 2\tilde{a}_{09})\,\psi_0^2 + 45\psi_0^4\big] \sum_q (\psi_{01q}^*\psi_{02q} + \psi_{02q}^*\psi_{01q} + \text{permutations})\,, \end{aligned} \tag{9}$$

where $V(\psi_0)$ is given by Eq. (4), with ψ_0 being a solution of Eq. (5) and $\dot{\psi}_{0jq} = (\partial/\partial t)\,\psi_{0jq}$.

These results are generalizations of those found by McMillan [6] and Ribeiro Filho [10], for the case of $\tilde{a}_{06} = 0$. It is worthwhile to stress that McMillan has omitted the contribution of the last term in Eq. (9), which we will retain because it is necessary in order to calculate the amplitude fluctuation modes or amplitudons. We can observe from Eqs. (8) and (9) that ψ_{0jq} and ψ^*_{0j-q} are coupled modes, and in this case, in order to avoid the crossed terms in amplitude and phase, we use the standard expression for phase (P_{jq}) and amplitude (A_{jq}) in terms of symmetric and anti-symmetric combinations: $A_{jq} = 1/\sqrt{2}\,(\psi_{0jq} + \psi^*_{0j-q})$; $A^*_{jq} = 1/\sqrt{2}\,(\psi^*_{0jq} + \psi_{0j-q}) = A_{j-q}$; $P_{jq} = 1/\sqrt{2}\,(\psi_{0jq} - \psi^*_{0j-q})$; and $P^*_{jq} = 1/\sqrt{2}\,(\psi^*_{0jq} - \psi_{0j-q}) = -P_{j-q}$. Substituting these expressions in Eqs. (8) and (9), we obtain:

$$T^+ = \sum_q \tfrac{1}{4} \sum_{i,j=1}^{3} T_{ij} \dot{A}^*_{iq} \dot{A}_{jq}\,, \tag{10}$$

$$\begin{aligned} V^+ = \tfrac{1}{2}\Bigg\{ &\sum_{jq} \left[\tfrac{1}{4}\,3\tilde{a}_{03}\psi_0 + \tfrac{1}{2}\,3\tilde{a}_{04}\psi_0^2 + \tfrac{1}{4}\,105\tilde{a}_{06}\psi_0^4 + \tilde{a}_{07}(\mathbf{Q}_j\cdot\mathbf{q})^2 + \tilde{a}_{08}(\mathbf{Q}_j\times\mathbf{q})^2\right] A^*_{jq}A_{jq} \\ &- \left[\tfrac{1}{4}\,3\tilde{a}_{03}\psi_0 - (3\tilde{a}_{04} + 2\tilde{a}_{09})\,\psi_0^2 - 45\tilde{a}_{06}\psi_0^4\right] \\ &\times \sum_q \left(A^*_{1q}A_{2q} + A^*_{2q}A_{1q} + \text{permutations}\right)\Bigg\}\,, \end{aligned} \tag{11}$$

$$T^- = \sum_q \tfrac{1}{4} \sum_{i,j=1}^{3} T_{ij} \dot{P}^*_{jq} \dot{P}_{jq}\,, \tag{12}$$

$$\begin{aligned} V^- = \tfrac{1}{2}\Bigg(&\sum_{jq} \left[\tfrac{1}{4}\,3\tilde{a}_{03}\psi_0 + \tilde{a}_{07}(\mathbf{Q}_j\cdot\mathbf{q})^2 + \tilde{a}_{08}(\mathbf{Q}_j\times\mathbf{q})^2\right] P^*_{jq}P_{jq} \\ &+ \tfrac{1}{4}\,3\tilde{a}_{03}\psi_0 \sum_q \left(P^*_{1q}P_{2q} + P^*_{2q}P_{1q} + \text{permutations}\right)\Bigg)\,, \end{aligned} \tag{13}$$

and we can write the Lagrangian as

$$L = L_0 + L^+ + L^- \tag{14}$$

such that $L^+ = T^+ - V^+$, $L^- = T^- - V^-$, and $L_0 = V(\psi_0)$.

The Euler-Lagrange equations obtained from Eq. (14) implicate two distinct systems of ordinary differential equations (in time), one for phasons calculations and another for amplitudons. The time-Fourier transform of these two systems of differential equations, for instance, $P_{jq} = (1/2\pi)\int \mathrm{d}\omega\, P_{jq\omega}\exp(i\omega t)$, leads to two systems of three homogeneous linear equations in $P_{jq\omega}$. The condition for the existence of non-trivial solutions is given by the secular equations $\mathrm{Det}\,(\omega^2 T - V) = 0$, where ω is the frequency, T is the matrix of elements T_{ij}, and the matrix elements of V are, for each system, given by

$$V_{ij} = \left(\frac{\partial^2 V^\pm}{\partial M^*_{iq}\,\partial M_{jq}}\right)_0\,, \tag{15}$$

where $V^+ \Rightarrow M \equiv A$ and $V^- \Rightarrow M \equiv P$, in the equation above. The secular equation solutions which represent the frequencies of fluctuation modes can be found applying the standard procedure in matrices $T = (T_{ij})$ and $V = (V_{ij})$, respectively. We can see that among these six modes, four are optical, that is, $\omega(q = 0) \neq 0$, three of them being amplitudons and one a phason. The other two are acoustic phasons. In order to write the expression of these modes, we will consider two distinct directions of **q**. We will assume **q** parallel to $\mathbf{Q}_1$ and, also, **q** perpendicular to $\mathbf{Q}_1$. In the first case we find the following amplitudons:

$$M^*\omega^2 = \tfrac{1}{2}\,3\tilde{a}_{03}\psi_0 - \left(\tfrac{1}{2}\,3\tilde{a}_{04} + 2\tilde{a}_{09}\right)\psi_0^2 - \tfrac{1}{4}\,75\tilde{a}_{06}\psi_0^4 + \tfrac{1}{4}\left(\tilde{a}_{07} + 3\tilde{a}_{08}\right)Q_1^2q^2 \ldots (E_{2g})\,, \tag{16}$$

$$M^*\omega^2 = \tfrac{1}{2}\,3\tilde{a}_{03}\psi_0 - \left(\tfrac{1}{2}\,3\tilde{a}_{04} + 2\tilde{a}_{09}\right)\psi_0^2 - \tfrac{1}{4}\,75\tilde{a}_{06}\psi_0^4 + \tfrac{1}{4}\left(3\tilde{a}_{07} + \tilde{a}_{08}\right)Q_1^2q^2 \ldots (E_{2g})\,, \tag{17}$$

$$M^*\omega^2 = -\tfrac{1}{4}\,3\tilde{a}_{03}\psi_0 + \left(\tfrac{1}{2}15\tilde{a}_{04} + 4\tilde{a}_{09}\right)\psi_0^2 + \tfrac{1}{4}\,465\tilde{a}_{06}\psi_0^4 + \tfrac{1}{2}\left(\tilde{a}_{07} + \tilde{a}_{08}\right)Q_1^2q^2 \ldots (A_{1g})\,; \tag{18}$$

and the following phasons:

$$M^*\omega^2 = \tfrac{1}{4}(\tilde{a}_{07} + 3\tilde{a}_{08})\,Q_1^2q^2 \ldots (E_{1u}), \tag{19}$$

$$M^*\omega^2 = \tfrac{1}{4}(3\tilde{a}_{07} + \tilde{a}_{08})\,Q_1^2q^2 \ldots (E_{1u}), \tag{20}$$

$$M^*\omega^2 = \tfrac{1}{4}9\tilde{a}_{03}\psi_0 + \tfrac{1}{2}(\tilde{a}_{07} + \tilde{a}_{08})\,Q_1^2q^2 \ldots (B_{1u})\,. \tag{21}$$

The normal modes can be classified in accordance with the irreducible representations of the symmetry group of the system. In Eqs. (16) to (21), E_{2g}, A_{1g}, E_{1u}, and B_{1u} are the symmetry character of the irreducible representations of the space group D_{6h}–6/mmm that characterizes the hexagonal structure of the incommensurate phase of 2H-$TaSe_2$. These results of $M^*\omega^2$, where ω is the mode frequency, are numerically sketched in Figs. 2 to 4. The value of M^* is 206 a.u. [6].

Equations (16) to (18) of amplitudons are completly new, due to the contribution of the sixth order term in those expressions. The phason expressions, given by Eqs. (19) and (20) are the two hydrodynamic modes obtained by other authors [6,10]. When we consider the case of **q** perpendicular to $\mathbf{Q}_1$ we get the following new expressions for the

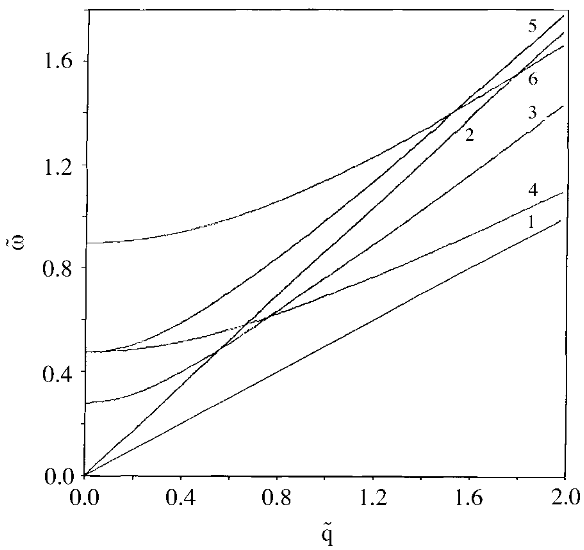

Fig. 2. Normal modes of the hexagonal incommensurate CDW, with $\tilde{\omega} = M^{*1/2}\omega$; $\tilde{q} = \tilde{a}_{07}^{1/2}Q_{1q}$; $T = 112.7$ K and $\tilde{a}_{06} = 1$. The curves correspond to (1) E_{1u}, (2) E_{1u}, (3) B_{1u}, (4) E_{2g}, (5) E_{2g}, and (6) A_{1g}

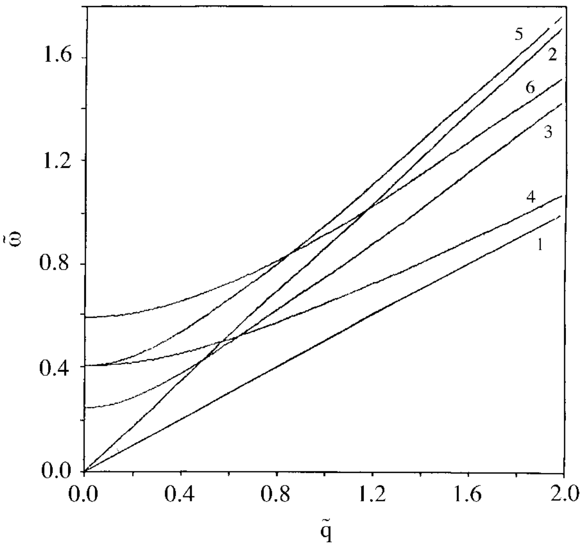

Fig. 3. Normal modes of the hexagonal incommensurate CDW, with $\tilde{\omega} = M^{*1/2}\omega$; $\tilde{q} = \tilde{a}_{07}^{1/2} Q_{1q}$; $T = 117.9\,\mathrm{K}$ and $\tilde{a}_{06} = 1$. The curves correspond to (1) E_{1u}, (2) E_{1u}, (3) B_{1u}, (4) E_{2g}, (5) E_{2g}, and (6) A_{1g}

three amplitudons (E_{2g}, E_{2g}, A_{1g}) and three phasons (E_{1u}, E_{1u}, B_{1u}), respectively:

$$M^*\omega^2 = \tfrac{1}{2}\, 3\tilde{a}_{03}\psi_0 - \left(\tfrac{1}{2}\, 3\tilde{a}_{04} + 2\tilde{a}_{09}\right)\psi_0^2 - \tfrac{1}{4}\, 75\tilde{a}_{06}\psi_0^4 + \tfrac{1}{4}\left(3\tilde{a}_{07} + \tilde{a}_{08}\right) Q_1^2 q^2 \ldots (E_{2g}), \quad (22)$$

$$M^*\omega^2 = \tfrac{1}{2}\, 3\tilde{a}_{03}\psi_0 - \left(\tfrac{1}{2}\, 3\tilde{a}_{04} + 2\tilde{a}_{09}\right)\psi_0^2 - \tfrac{1}{4}\, 75\tilde{a}_{06}\psi_0^4 + \tfrac{1}{4}\left(\tilde{a}_{07} + 3\tilde{a}_{08}\right) Q_1^2 q^2 \ldots (E_{2g}), \quad (23)$$

$$M^*\omega^2 = -\tfrac{1}{4}\, 3\tilde{a}_{03}\psi_0 + \left(\tfrac{1}{2} 15\tilde{a}_{04} + 4\tilde{a}_{09}\right)\psi_0^2 + \tfrac{1}{4}\, 465\tilde{a}_{06}\psi_0^4 + \tfrac{1}{2}\left(\tilde{a}_{07} + \tilde{a}_{08}\right) Q_1^2 q^2 \ldots (A_{1g}), \quad (24)$$

$$M^*\omega^2 = \tfrac{1}{4}\left(3\tilde{a}_{07} + \tilde{a}_{08}\right) Q_1^2 q^2 \ldots (E_{1u}), \quad (25)$$

$$M^*\omega^2 = \tfrac{1}{4}\left(\tilde{a}_{07} + 3\tilde{a}_{08}\right) Q_1^2 q^2 \ldots (E_{1u}), \quad (26)$$

$$M^*\omega^2 = \tfrac{1}{4}\, 9\tilde{a}_{03}\psi_0 + \tfrac{1}{2}\left(\tilde{a}_{07} + \tilde{a}_{08}\right) Q_1^2 q^2 \ldots (B_{1u}). \quad (27)$$

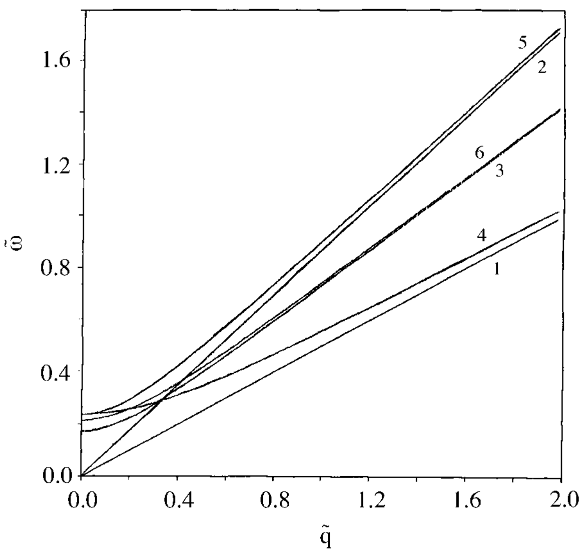

Fig. 4. Normal modes of the hexagonal incommensurate CDW, with $\tilde{\omega} = M^{*1/2}\omega$; $\tilde{q} = \tilde{a}_{07}^{1/2} Q_{1q}$; $T = 121.5\,\mathrm{K}$ and $\tilde{a}_{06} = 1$. The curves correspond to (1) E_{1u}, (2) E_{1u}, (3) B_{1u}, (4) E_{2g}, (5) E_{2g}, and (6) A_{1g}

4. Numerical Results

In this section we present the results of numerical calculations of normal mode frequencies (phasons and amplitudons) for the triple charge-density-wave hexagonal incommensurate superlattice of a single layer of the compound 2H-$TaSe_2$. We have obtained the mode frequencies in terms of the wave-vector $\mathbf{q}$, as well as the behaviour of these modes for different values of temperature in the range of 112 to 122 K, for $\tilde{a}_{06} = 1$. Figs. 2 to 4 give the pictures when we consider the case $\tilde{a}_{06} = 1$ and temperatures T of 112.7, 117.9 and 121.5 K. We have used the same values introduced by other authors [10,11] for the phenomenological parameters, that is, we have taken $\tilde{a}_{03} = \frac{4}{30}$, $\tilde{a}_{04} = \frac{8}{3}$ and $\tilde{a}_{09} = -3.9$, by virtue of the new generalized free-energy expression (9). These figures are sketched from the Eqs. (16) to (21) with $\tilde{a}_{08} = 0$ and the coordinates expressed by $\tilde{\omega} = M^{*1/2}\omega$ and $\tilde{q} = \tilde{a}_{07}^{1/2} Q_{1q}$. In this numerical illustration we have used the Landau parameter [9] $\tilde{a}_{020} = 2.35 \times 10^{-4}$ (SI). We can observe that the acoustic phasons are similar to those obtained by other authors [6,10] indicating that the inclusion of the sixth-order term in the homogeneous part of the free-energy potential changes only the expressions of the optical amplitudons.

5. Conclusions

The calculations that describe excitations of the lattice below a phase transformation from a normal structure to an incommensurate phase is not straightforward due to the loss of the translational symmetry. In spite of this evidence, it is necessary to get information about incommensurate compounds like 2H-$TaSe_2$. Our numerical work has concentrated in simulating this material, using the generalized expression of the Landau-Lifshitz free energy [9], assuming phenomenological parameters in order to get some pictures of normal-mode frequencies of the hexagonal incommensurate phase as the temperature changes. The Lagrangian mechanics background has been used in order to get our analytical expressions. We must stress that the parameter values used in this work are in good agreement with experimental data [12]. The eigenmodes frequencies are calculated using the above mentioned free energy, where we have considered contributions up to sixth-order term in the homogeneous part of the termodynamical potential, such that we have got new results, which are consistent with those obtained by other authors [6,10] when we consider $\tilde{a}_{06} = 0$. It is worth to emphasize that only phasons expressions remain the same despite the inclusion of mentioned sixth-order term. We have considered only intralayer contributions in the free-energy expression; and in the case of the hexagonal incommensurate phase, the cubic ("lock-in") term in the generalized Landau-Lifshitz free energy drops out. In order to compute the eigenmode frequency expressions, we have considered two possibilities for the $\mathbf{q}$-directions: parallel and perpendicular to $\mathbf{Q}_1$. McMillan (1975) [6] has shown that phason modes involve long-wavelength distortions of the charge-density-lattice. The comparison of these results with those of other authors, despite different contributions from our new equations, maintain the fundamental characteristics of the pictures involving the phasons and amplitudons, that is, we can clearly observe variations of the energy gaps for the optical modes (E_{2g}, A_{1g}, and B_{1u}). From analytical expressions and Figs. 2 to 4, we see that four modes are optical, where three are amplitudons and one is phason, and the two other are acoustic phasons.

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References

[1] L. M. Floría and J. J. Mazo, Adv. Phys. **45**, 505 (1996).
[2] G. Grüner, Rev. Mod. Phys. **60**, 1129 (1988).
[3] J. A. Wilson and A. D. Yoffe, Adv. Phys. **18**, 193 (1969).
[4] Yu. I. Latyshev, O. Laborde, P. Monceau, and S. Klaumünzer, Phys. Rev. Lett. **78**, 919 (1997).
[5] G. Montambaux, Phys. Rev. B **54**, R17273 (1996).
[6] W. L. McMillan, Phys. Rev. B **12**, 1187 (1975); **14**, 1494 (1976); **16**, 543 (1977).
A. E. Jacobs and M. B. Walker, Phys. Rev. B **26**, 206 (1982).
[7] C. H. Chen, J. M. Gibson, and R. M. Fleming, Phys. Rev. B **26**, 184 (1982); Phys. Rev. Lett. **47**, 723 (1981).
[8] M. O. Steinitz and J. G. Genossar, Solid State Commun. **29**, 519 (1979).
[9] J. F. M. Rocha, D. S. de Vasconcelos, and A. Ribeiro Filho, phys. stat. sol. (b) **205**, 473 (1998).
S. A. Jackson, P. A. Lee, and T. M. Rice, Phys. Rev. B **17**, 3611 (1978).
[10] A. Ribeiro Filho, Rev. Bras. de Física (now Braz. J. Phys.) **16**, 233 (1986).
[11] K. Nakanishi and H. Shiba, J. Phys. Soc. Japan **43**, 1839 (1977); **44**, 1965 (1978).
[12] E. F. Steigmeier, G. Harbeke, H. Auderset, and F. Di Salvo, Solid State Commun. **20**, 667 (1976).