Raman spectroscopy of the charge-density-wave state in $1T$- and $2H$-$TaSe_2$

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The Raman spectrum of $1T$-$TaSe_2$ has been studied for $2 < T < 480$ K. Changes in the Raman spectra have been observed which are correlated with the 473-K phase transition from the commensurate charge-density-wave (CDW) state to the incommensurate CDW state. The temperature dependence of the Raman spectra is related to changes in the magnitude of the CDW order parameter in the commensurate state. Comparison of the temperature-dependent Raman spectra of $1T$- and $2H$-$TaSe_2$, suggests that the observation of multiple CDW-induced lines at energies below 100 cm^{-1} is partly due to the existence of secondary order parameters in the CDW state.

INTRODUCTION

Raman spectroscopy has been a useful tool in the study of systems showing charge-density-wave (CDW) states. Smith *et al.*[1] have shown that the CDW-induced collapse of the Brillouin-zone results in the appearance of many additional phonon lines in the Raman spectra. In addition, Holy *et al.*[2] have shown these new modes can be used to obtain information about the symmetry of the commensurate lattice distortion. Tsang *et al.*[3] and Steigmeier *et al.*[4] have shown that studies of the temperature dependence of the Raman spectra near the CDW phase transitions are useful for understanding the dynamics of the transition to the charge-density-wave state.

In this paper we report on the temperature-dependent Raman spectra of the transition-metal dichalcogenide compound $1T$-$TaSe_2$ and compare our results with new and previously published results on the Raman spectra of $2H$-$TaSe_2$. We find that the commensurate CDW phase Raman spectra of $1T$-$TaSe_2$ and $2H$-$TaSe_2$ are very similar. Both show temperature-dependent modes whose frequencies follow the temperature dependence of the CDW order parameter as determined from independent measurements. Similarities in the temperature dependence of the intensities and linewidths of these modes have also been observed near the corresponding CDW phase transitions in both materials. The similarities in the behavior of the Raman lines and the substantial differences in the Fermi surfaces and CDW lattice distortions of these two compounds allow us to draw a number of conclusions regarding the character of the commensurate CDW state. For example, Steigmeier *et al.*[4] pointed out that the two different mechanisms proposed for the CDW instability in $2H$-$TaSe_2$, the nesting mechanism and the saddle-point mechanism can have consequences as to the interaction among the multitude of $k=0$ CDW excitations in the commensurate phase. We show that the similarity of the Raman spectra of $2H$- and $1T$-$TaSe_2$ in their respective commensurate CDW states suggests that the driving mechanism for the CDW instability is the same in both cases. In addition, we confirm that substantial changes in the Raman spectra of CDW systems occur at the first-order commensurate-state to incommensurate-state CDW transition. Finally, we find that our results are consistent with models of the charge-density-wave state involving more than one order parameter or coupled order parameters.[5,6] We show that this situation can arise in two different ways: through the existence of higher spatial harmonics of the fundamental CDW distortion[4-6] and through an explicit consideration of the dependence of the free energy of the CDW on the amplitude of the lattice distortion and the charge modulation.

The $1T$ and $2H$ polytypes of $TaSe_2$ are closely related. The $1T$ polytype is metastable at room temperature; it is prepared by rapidly quenching $1T$-$TaSe_2$ from high temperatures. The $2H$ material used was grown at 700 °C and then cooled. The $1T$ polytype consists of a single $TaSe_2$ formula per unit cell, with the selenium ions octahedrally coordinated about the tantalum ions, while the $2H$ polytype contains two $TaSe_2$ formulas per unit cell, with trigonal prismatic coordination. This difference in the crystal structure is accompanied by substantial changes in the band structure, the Fermi surface and the phonon dispersion. While both materials show CDW instabilities, the respective instabilities differ considerably in detail.[1,2] In $2H$-$TaSe_2$ a transition from the undis-

torted state to the incommensurate CDW state occurs on cooling at 122 K followed by a first-order transition to a commensurate CDW state for $T > 90$ K (depending on the sample) where the in-plane lattice constant is tripled. The transition from the $1T$ to another polytype occurs at 600 K, a temperature below that of the incommensurate CDW to normal-state transition in $1T$-$TaSe_2$. The first-order transition from the incommensurate to the commensurate CDW state at 473 K in $1T$-$TaSe_2$ results in a new unit cell rotated 13.9 deg from the original unit cell and containing 13 tantalum and 26 selenium ions. These differences should be reflected in the Raman spectra of the respective commensurate CDW phases.

EXPERIMENTAL DETAILS AND RESULTS

All of the measurements described in this paper were made on cleaved single-crystal samples. The spectra were obtained using different lines from Ar-ion, Kr-ion, and dye lasers. The scattered light was analyzed with a double monochromator and detected with photon-counting electronics. The samples were measured either immersed in liquid He at 2 K or attached to a copper heat sink in an evacuated chamber. In the latter situation, the temperature was measured with a thermocouple.

In Fig. 1 we show the Raman spectra of $1T$-$TaSe_2$ at several temperatures in the range 2–480 K. No data are available for the undistorted state of $1T$-$TaSe_2$ because of the interpolytype phase transition.[7] We will discuss the spectra in Fig. 1 in two parts: first the lines which occur above 100 cm^{-1} and then those which occur below 100 cm^{-1}. The Raman spectra of $1T$-$TaSe_2$ in the absence of a lattice distortion should consist of a pair of lines of A_{1g} and E_g symmetry. Infrared studies by Lucovsky *et al.*[8] on $1T$-$TaSe_2$ and related compounds indicate that these lines should occur between 150 and 250 cm^{-1}. We identify the Raman lines seen in Fig. 1 between 150 and 200 cm^{-1} as arising from the normal Raman-active phonons of $1T$-$TaSe_2$ and from new Raman-active phonons due to the folding of the optical-phonon dispersion curves in the presence of a commensurate superlattice. These lines show little shift in frequency with temperature over the temperature range studied. The intensity of these lines decreases abruptly at the first-order commensurate to incommensurate CDW phase transition. They are not observable in the incommensurate CDW phase and resemble our results in $1T$-TaS_2.[1]

The spectral features in Fig. 1 observed below 100 cm^{-1} show different behavior. The two strongest lines for $T < 473$ K have A_{1g}-like symmetry and shift to lower energies as the commensurate to incommensurate CDW transition is approached. In contrast, the weaker line at 83 cm^{-1} shows E_g-like symmetry and no appreciable shift in energy as a function of temperature for $T < 473$ K. The peak intensity of the higher-lying A_{1g}-like symmetry line (99 cm^{-1} at 2 K) approaches zero at the first-order phase transition from the commensurate to the incommensurate CDW state. As a result, in $1T$-$TaSe_2$ near the 473-K commensurate-to-incommensurate transition, the Raman spectra are dominated by the lower energy A_{1g}-like symmetry mode. Above the transition, the scattering is characterized by a broad continuum with a peak below 50 cm^{-1}. Similar results, i.e., an A_{1g} symmetry mode which both softens and weakens as the commensurate-incommensurate transition is approached from below and a broad continuum above the transition, have been observed in $2H$-$TaSe_2$.[3,4]

Hughes and Pollak[9] have measured x-ray photo-

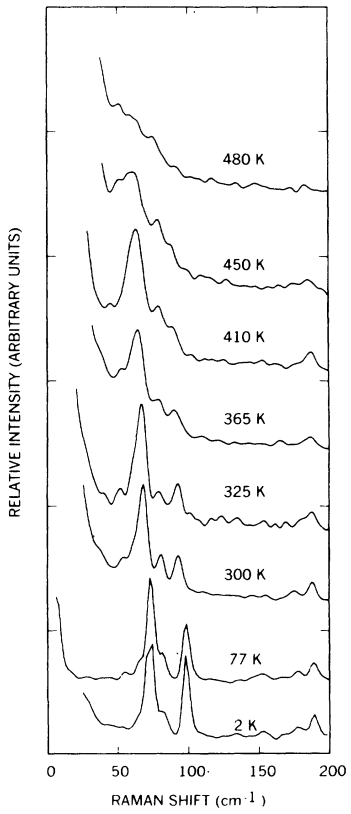

FIG. 1. Raman spectra of $1T$-$TaSe_2$ at 2, 77, 300, 365, 410, 450, and 480 K. The spectra were excited using the 5145-Å line of a Ar-ion laser. T_{comm} in $1T$-$TaSe_2$ is at 473 K. The modes at 72 and 99 cm^{-1} have A_{1g}-like symmetry while the mode at 83 cm^{-1} has E_g-type symmetry.

emission spectra of $1T$-$TaSe_2$ as a function of temperature between 10 and 300 K. They observed that x-ray photoemission from the $4f$ bound states of the Ta ions shows several discrete lines and have attributed this to the existence of inequivalent Ta sites in the commensurate CDW state. The splitting between these lines decreases with increasing temperature. They concluded that the magnitude of the splitting is proportional to the amplitude of the modulation of the conduction electron density in the commensurate CDW state. In Fig. 2, we plot the temperature dependence of a number of the Raman lines shown in Fig. 1. We find the Raman lines can be divided into two groups, those which show softening at the commensurate-to-incommensurate CDW phase transition and those which show only a very weak temperature dependence. In Fig. 2, we also plot the temperature dependence of the Ta $4f$ splitting as measured by Hughes and Pollak, normalized to the 2-K energy of the lower of our two A_{1g} symmetry lines observed below 100 cm^{-1}. As can be seen, the temperature dependences of the Ta $4f$ splitting and of the Raman frequency of this line are similar. The temperature dependence of the frequency of the higher-lying A_{1g} mode is also similar to the temperature dependence of the splitting of the emission from the Ta $4f$ levels. This shows that the frequencies of these lines reflect the magnitude of the commensurate CDW distortion. A similar conclusion follows from the observation that the temperature dependence of the frequency of a number of low-lying modes in the Raman spectra of $2H$-$TaSe_2$ in the commensurate CDW state is similar to the temperature dependence of the intensity of the superlattice spots observed by elastic neutron scattering.[3-5]

In Fig. 1, we find that the incommensurate-to-commensurate CDW phase transition produces changes in the strengths of some of the low-lying CDW-induced modes of $1T$-$TaSe_2$. In Fig. 3, we show the temperature dependence of the peak strengths of the two low-lying A_{1g}-like symmetry modes observed in $1T$-$TaSe_2$.[10] The intensities of the low-lying modes are normalized against the intensity of the symmetry allowed Raman mode at 190 cm^{-1}. (We assume that the 190-cm^{-1} A_{1g} mode is one of the two Raman-allowed modes of the undistorted lattice and expect that its strength should not change appreciably for temperatures below the first-order commensurate-to-incommensurate CDW phase transition.) We find that the relative peak strength of the low-lying A_{1g} mode shows only a weak variation with temperature below the commensurate-to-incommensurate CDW transition. In contrast, as was suggested in Fig. 1, the relative peak strength of the 99-cm^{-1} A_{1g} mode

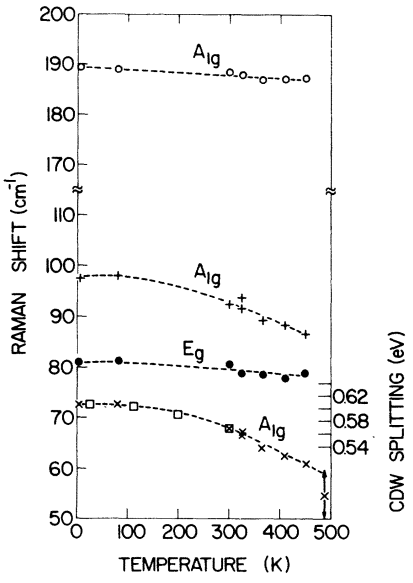

FIG. 2. Temperature dependence of the frequency shifts of four lines in the Raman spectra of $1T$-$TaSe_2$. The labels refer to the polarization properties of these lines. The square boxes are obtained from the photoemission results of Hughes and Pollak (Ref. 9) on the splitting of the emission from the Ta $4f$ levels and refer to the right-hand scale. The other points are obtained from Raman-scattering measurements and refer to the left-hand scale. Note the break in the ordinate scale.

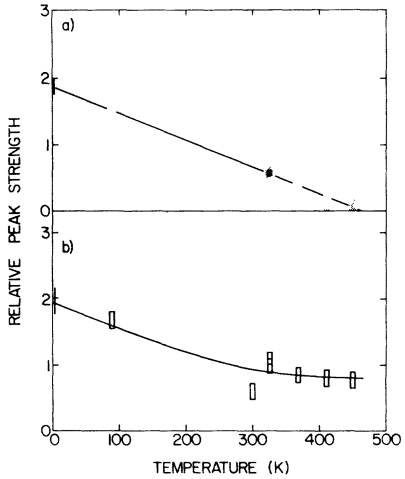

FIG. 3. (a) Temperature dependence of the peak strength of the 99-cm^{-1} (at 2 K) Raman mode in $1T$-$TaSe_2$. (b) Temperature dependence of the peak strength of the 72-cm^{-1} (at 2 K) Raman mode in $1T$-$TaSe_2$. In both figures, the experimental data have been normalized against the intensity of the 189-cm^{-1} A_{1g} mode and both the frequency and Bose factors in the expression for the Raman cross section removed (see Ref. 10).

decreases rapidly with increasing temperature, approaching zero at 473 K. A similar temperature dependence is observed in $2H$-$TaSe_2$. In Fig. 4, we plot the relative peak strength (with the intensity normalized against the intensity of the 234-cm^{-1} A_{1g} mode) of the 82-cm^{-1} A_{1g} mode in $2H$-$TaSe_2$ and show that it undergoes a similar decline with increasing temperature below the commensurate-to-incommensurate phase transition in $2H$-$TaSe_2$. We find, therefore, that the first-order commensurate-to-incommensurate CDW phase transition in both $1T$- and $2H$-$TaSe_2$ is associated with the gradual disappearance of an A_{1g} Raman mode near 100 cm^{-1}.

In Figs. 5 and 6, we plot the full width at half-maximum of the 82- and 99-cm^{-1} A_{1g} lines in $2H$-$TaSe_2$ and $1T$-$TaSe_2$, respectively. Both lines increase in width as the commensurate to incommensurate CDW phase transition is approached. Our results are obtained in the $z(xx)z$ scattering geometry and are considerably narrower than the results obtained by Steigmeier *et al.*[4] in $2H$-$TaSe_2$ for temperatures above 50 K. Steigmeier *et al.* measure a low-temperature linewidth comparable to the low-temperature linewidth we observe. However, above 50 K, they obtain a value for the linewidth which increases rapidly with increasing temperature. At 55 K, their full width at half-maximum for this line is 11 cm^{-1} and at 65 K, it is almost 16 cm^{-1}. This difference may reflect differences in the samples used. The $2H$-$TaSe_2$ samples used in this experiment were from the same batch used by Craven and Meyer[11] and Holy *et al.*[2] and have higher transition temperatures (100–110 K) for the commensurate to incommensurate CDW phase transition than the samples used by Steigmeier *et al.* (90 K).

DISCUSSION

The temperature-dependent Raman spectra of $1T$- and $2H$-$TaSe_2$ in the commensurate CDW states show strong qualitative similarities, especially if the temperature is normalized to the appropriate commensurate-to-incommensurate phase-transition temperature. Both systems show several temperature-dependent excited states of the CDW ground state. These multiple CDW-induced modes have been attributed to several different causes.[4] These causes include (i) a symmetry-induced splitting of the degenerate amplitude and phase modes of the CDW state, (ii) the coupling of Σ_1 and Σ_3 symmetry phonons to the longitudinal charge fluctuations of the CDW, and (iii) the existence of secondary order parameters in the free energy of the CDW state. In this section, we show

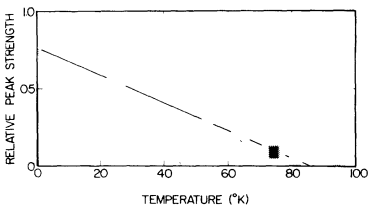

FIG. 4. Temperature dependence of the peak strength of the 82-cm^{-1} (at 2 K) Raman mode in $2H$-$TaSe_2$. As in Fig. 3, the experimental data have been normalized against the strength of one of the symmetry allowed A_{1g} phonons of undistorted $2H$-$TaSe_2$.

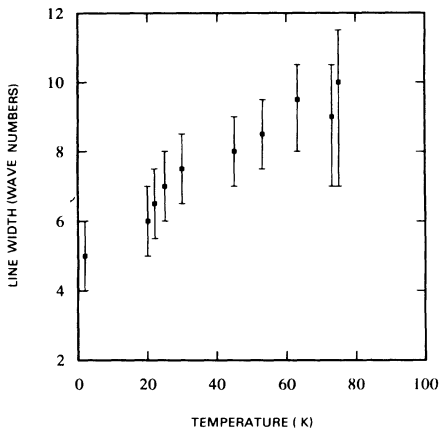

FIG. 5. Temperature dependence of the linewidth at half-maxima of the 82-cm^{-1} A_{1g} mode in $2H$-$TaSe_2$.

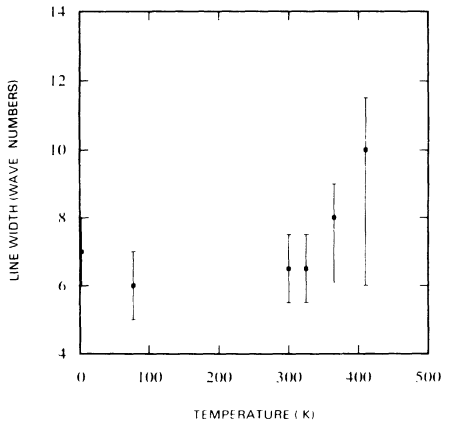

FIG. 6. Temperature dependence of the linewidth at half-maximum of the 99-cm^{-1} A_{1g} mode in $1T$-$TaSe_2$.

that our results on $1T$- and $2H$-$TaSe_2$ shed some light on each of these possibilities. In addition, the CDW-induced Raman spectra of $1T$- and $2H$-$TaSe_2$ can also provide some information on whether the CDW transition in $2H$-$TaSe_2$ is due to a nesting or a saddle-point Fermi-surface instability.

The elementary excitations of a one-dimensional CDW are a phase and an amplitude mode. In the presence of an interaction between the three distinct one-dimensional charge-density waves present in these hexagonal systems, one expects a symmetry induced splitting of the threefold degenerate phase and amplitude modes. This would result in several temperature-dependent low-energy Raman lines. Holy *et al.*[2] treated the problem of commensurate $2H$-$TaSe_2$ in the limit where the CDW distortion can be expressed as a sum of six equivalent Σ_1 symmetry phonons from the first Brillouin zone. Holy *et al.* showed that one expected on the basis of group-theoretical considerations A_{1g} and E_{2g} symmetry amplitude and phase modes.

It has been shown however that the energies of the amplitude modes should not be strongly effected by the phase transition from the incommensurate to the commensurate CDW state while the energies of the phase modes should be considerably reduced at this transition.[12] Our results and the results of Steigmeier *et al.*[4] show a single low-energy Raman line which only broadens at the incommensurate to commensurate CDW phase transition with no substantial change in energy, while the higher-lying A_{1g} mode disappears. This behavior is surprising given the symmetry based assignment of Holy *et al.* in which the lowest-energy A_{1g} and E_{2g} modes are called phasons and the higher-energy mode an amplitude mode.[2] While it is possible to reverse the assignment of Holy *et al.*, the following argument indicates this also does not produce a likely situation.

Lee, Rice, and Anderson[13] have considered the dynamics of a one-dimensional linear chain conductor. They have shown that the energy $\omega(0)$ of the one-dimensional amplitude mode in the incommensurate CDW state at $T=0$ is $\omega(0)=(1.5\Lambda)^{0.5}\omega_0$, where Λ is the electron-phonon coupling constant and ω_0 is the unscreened phonon energy. They have also shown that the energy ω_T of the long-wavelength phase mode in the commensurate CDW state is $\omega_T = M(\Delta/W)^{M/2-1}\omega(0)/1.5$, where Δ is the energy of the CDW-induced gap, W is the width of the conduction band, and M is the commensurability factor ($M=3$ in $2H$-$TaSe_2$). Since the value of the CDW gap is determined by the temperature of the transition from the normal to the incommensurate CDW state and the bandwidth is greater than 2 eV, the energy of the one-dimensional phase mode in the commensurate CDW state will be less than the energy of the amplitude mode. The symmetry-induced splitting of the threefold degeneracy of the amplitude and phase modes should not effect this result. Therefore, the temperature dependence, symmetry properties, and energies of the low-energy A_{1g} modes taken together indicate that the simple model used by Holy *et al.*[2] requires some refinement.

The group-theoretical results of Holy *et al.*[2] were based on a model which only couples Σ_1 symmetry phonons. Steigmeier *et al.*[4] have suggested that additional CDW induced modes can arise from the fact that the phonons along the Σ axis do not have well defined longitudinal and transverse branches. As a result, electrons and holes scattering across the Fermi surface can couple to both phonon branches. However, Moncton *et al.*[5] have found that the displacement pattern of the commensurate CDW state has Σ_1 symmetry. This makes it unlikely therefore that the additional CDW-dependent lines observed in $1T$- and $2H$-$TaSe_2$ are due to the interaction of carriers at the Fermi surface with both Σ_1 and Σ_3 symmetry phonons, which we called model (ii).

If the free energy of the commensurate CDW state is described by

$$F_0 = A(q, T)\gamma^2 + B\gamma^3 + C\gamma^4 + \dots ,$$

where γ is the order parameter of the phase transition, then the CDW state will be characterized by a single soft mode. However, neutron-scattering results of Moncton *et al.*[5] show that the free energy of the commensurate CDW state is somewhat more complicated. There is the possibility of both secondary and coupled order parameters. Such secondary or coupled order parameters will produce extra CDW-dependent modes in the Raman spectra of the commensurate CDW state.

The commensurate CDW displacement pattern derived by Moncton *et al.* implies coupling between the Σ_1 symmetry optical and acoustical phonons and the longitudinal charge fluctuations. Since it can also be shown that the amplitude of the CDW distortion is proportional to the magnitude of the CDW charge modulation, the free energy of the commensurate CDW state must take the form

$$F' = A_0(q, T)\gamma_1^2 + A_1(q, T)\gamma_2^2 + A_2(q, T)\gamma_3^2 + B\gamma_1^3 + C\gamma_1^4 + e\gamma_1\gamma_2 + f\gamma_1\gamma_3 + \dots .$$

The linear coupling of the different γ_i's is necessary to obtain the linear relationship between the charge-density modulation and the lattice distortion. In this expression, only one $A_i(q, T)$ need go to zero at the transition temperature for a phase transition to occur. γ_1 can represent the charge

density while γ_2 and γ_3 represent the contributions of the acoustical- and optical-phonon displacement patterns to the CDW distortion.

Moncton *et al.* have also observed the existence of secondary lattice distortions in the incommensurate CDW state of $2H$-$TaSe_2$. This secondary distortion corresponds to a spatial harmonic of the fundamental CDW modulation. Bak and Emery[14] have shown that such higher harmonics are essential to the stabilization of the commensurate CDW state since the transition to the commensurate state is driven by an anharmonic term in the free energy. In the commensurate charge-density-wave state, higher spatial harmonics of the lattice distortion have no physical meaning. However, higher spatial harmonics of the charge-density wave can have a physical meaning since the charge density is defined both on the lattice sites and between the lattice sites. In the commensurate CDW state, the presence of higher harmonics in the charge distribution can change the amount of charge localized on and between the metal ion sites and thereby reduce the Coulomb energy of the commensurate state. As a result, the free energy of the commensurate state will depend on terms describing the magnitude of the spatial harmonic of the fundamental CDW charge distribution. Steigmeier *et al.*[4] have suggested that the multiple lines observed in the CDW spectra may arise from this reason.

The frequency of the A_{1g} mode which disappears at the commensurate to incommensurate CDW transition in $2H$-$TaSe_2$ is very close to the frequency of the Σ_1-symmetry LA phonon measured by Moncton *et al.*[5] in the incommensurate CDW state. As a result, the 82-cm^{-1} Raman line in $2H$-$TaSe_2$ may represent scattering from a finite wave-vector LA phonon. The momentum-conservation-selection rule for Raman scattering would be satisfied by the existence of the commensurate CDW superlattice. Moncton has pointed out that the temperature dependence of the frequency of this acoustical mode is surprising since it does not go soft at the 122-K CDW phase transition as expected given its symmetry and simple theoretical treatments of the CDW state. This acoustical-phonon mode would contribute strongly to the Raman scattering in the commensurate CDW state since it couples to longitudinal charge fluctuations. One would therefore expect the Raman spectra to show a soft mode associated with the CDW phase transition and a strongly temperature-dependent mode corresponding to this phonon branch. A quantitative treatment of this coupled mode picture is however not currently available so a detailed comparison is not possible. No quantitative treatment of the Raman scattering from the secondary charge-distortion model exists either. The experimental observation that the low-energy CDW induced lines are of comparable intensities appears however to be inconsistent with a secondary distortion model where one expects the amplitudes of the harmonics to be smaller than the amplitude of the fundamental. We therefore believe that the multiple-line commensurate CDW-state Raman spectra we observe in $2H$-$TaSe_2$ is related to the anomalous behavior of the Σ_1 symmetry acoustical phonon observed by Moncton *et al.*[5] The similarity in the appearance of the $1T$- and $2H$-$TaSe_2$ Raman spectra in their respective commensurate CDW states would then suggest that similar anomalous behavior would be associated with the transition from the normal state of $1T$-$TaSe_2$ to the incommensurate CDW state.

Steigmeier *et al.* suggested that the CDW-induced Raman spectra arising from a nesting Fermi-surface instability would qualitatively differ from the spectra arising from a saddle-point Fermi-surface instability.[4] In the simple nesting picture, each piece of Fermi surface only couples to one other piece of Fermi surface resulting in three one-dimensional charge-density waves. The three equivalent one-dimensional CDW's are weakly coupled to give the observed two-dimensional CDW. The symmetry induced splitting of the amplitude and phase modes will be relatively small in this case. In contrast, in the saddle-point picture, the two-dimensional aspect of the problem must be introduced at the outset as each of the six saddle points in the band structure will be coupled to two other saddle points. As a result, the symmetry-induced splitting of the CDW modes will be comparable to the energies of these modes. This point has been made explicitly by Tosatti.[15] The band structure of $1T$-$TaSe_2$ is consistent only with a nesting Fermi-surface instability and we observe that any splitting of our strong A_{1g}-like mode at 72 cm^{-1} and an E_g-like mode must be small. Similarly, we find that the splitting of the A_{1g} and E_{2g} symmetry modes in $2H$-$TaSe_1$ is also small compared to the energy of these lines (7 cm^{-1} out of 49 cm^{-1}) and this therefore suggests that the CDW in $2H$-$TaSe_2$ is driven by a nesting instability.

In conclusion, we have studied the temperature dependent Raman spectra of $1T$- and $2H$-$TaSe_2$. We have exploited the similarities in the crystal structures of these materials and the differences in the CDW transition temperatures to obtain a better understanding of the commensurate charge-density-wave state and its excitations. Comparison of the Raman spectra of these two compounds indicates that a saddle-point Fermi-surface instability is not necessary to understand the observation of multiple CDW lines in $2H$-$TaSe_2$. It also

indicates that the simple treatment of the commensurate charge-density wave characterized by only a single-order parameter describing the amplitude of the lattice distortion cannot explain the detailed behavior of the Raman spectra.

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