Proximity to a commensurate charge modulation in IrTe$_{2-x}$Se$_x$ ($x = 0$ and 0.45) revealed by Raman spectroscopy

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Received 3 July 2014, revised 9 August 2014
Accepted for publication 14 August 2014
Published 30 September 2014

Abstract

Optical phonons are employed to probe a structural phase transition of IrTe$_{2-x}$Se$_x$ at $T_C \sim 280$ K for $x = 0$ and $T_C \sim 491$ K for $x = 0.45$, which is dictated by Ir dimerization with charge modulations. Phonon Raman spectra show a discontinuous change at $T_C$ and thermal hysteresis. A $165$ cm$^{-1} A_g$ mode involving out-of-plane motions of Te atoms is mostly affected by Se doping. This highlights the role of interlayer Te–Te interactions in inducing the structural instability. For temperatures below $T_C$ both $x = 0$ and $x = 0.45$ compounds exhibit commonly five zone-folded peaks for the $128$ cm$^{-1} E_g$ in-plane mode and six zone-folded peaks for the $165$ cm$^{-1} A_g$ mode. This is taken as a signature of a respective quintupling and sextupling of the unit cell. Our results, on the one hand, are consistent with a 1/5 charge modulation reported by previous studies and, on the other hand, indicate that the ground state of IrTe$_{2-x}$Se$_x$ lies in proximity to a 1/6 commensurate charge modulation phase.

Keywords: charge density wave, structural phase transition, superstructure
1. Introduction

Transition metal dichalcogenides MX$_2$ (M = transition metal and X = chalcogen) have been widely studied for electronic instabilities toward broken symmetry such as superconductivity, charge density wave (CDW), and charge and orbital order [1–3]. This is owed to their quasi-two-dimensional electron structure, which is formed by strong intralayer covalent bonds and weak interlayer interactions. MX$_2$ is furthermore characterized by a high in-plane stiffness as a positively charged plane of M atoms is sandwiched between two planes of negatively charged X atoms [4]. In the present study, we focus on IrTe$_2$, which shows unconventional CDW and superconductivity [5–15].

IrTe$_2$ crystallizes in a trigonal CdI$_2$-type structure (space group Pm$ar{3}$m) at room temperature [4]. A layered metal is formed by edge-sharing IrTe$_6$ octahedra, which are stacked along the c-axis. This compound undergoes a structural transition to a triclinic phase (space group P$ar{1}$) at $T_C \sim 280$ K, accompanied by anomalies in electrical resistivity and magnetic susceptibility [11]. This is attributed to the formation of a CDW as in other dichalcogenides [13]. For temperatures below $T_C$, structural modulations are characterized by a $\vec{q} = 1/5(1, 0, 1)$ superstructure. Upon Se doping, $T_C$ increases up to 560 K while the structural modulation transforms from $\vec{q} = 1/5(1, 0, 1)$ to $\vec{q} = 1/6(1, 0, 1)$ [14].

Fermi surface nesting has originally been suggested as the driving force of the charge modulation. However, angle-resolved photoemission spectroscopy and optical measurements show the absence of a CDW gap [14–17]. Furthermore, the structural modulation is highly nonsinusoidal [14]. This calls into question the relevance of a conventional CDW scenario for IrTe$_2$. To resolve this enigma, a number of alternative mechanisms have been proposed—for example, orbital degeneracy of Ir $d_5$ and/or Te $p_5$ states, charge and orbital waves on the Ir sites, and a depolymerization-polymerization of interlayer Te bonds [14–17].

Very recently, single-crystal x-ray and neutron diffraction, Ir $L_3$-edge extended x-ray absorption fine structure, and scanning tunneling microscopy (STM) studies along with first-principles calculations [18–23] have revealed an important aspect of the structural modulations. They provide evidence for a partial formation of Ir–Ir and Te–Te dimers as well as for charge disproportionation among the dimerized and nondimerized Ir atoms assisted by the interaction between the Ir 5$d$ and Te 5$p$ orbitals. This has multiple consequences: (i) a switching of the conducting planes from the basal plane of the high-$T$ trigonal phase to a tilted plane normal to $\vec{q}$ of the low-$T$ triclinic phase, (ii) an enhancement of two-dimensionality, and (iii) a site-dependent depletion of density of states around the Fermi level, $E_F$.

In addition, resistivity and STM measurements show a second hysteretic transition at $T_S \sim 180$ K [22]. This is ascribed to a partial melting of a soliton lattice, giving rise to short-range charge modulations described by $q_{n} = (3n + 2)^{-1}$ ($n$: natural number). In this view, the 1/5 modulation corresponds to a periodic arrangement of two types of fundamental units, a 2$a^*$ soliton lattice and 3$a^*$ stripes, where $a^* = a \sin 60^\circ$ is the intercolumn spacing and $a$ is the lattice constant. Although the periodic Ir and Te dimer stripes have been established as key ingredients for the structural phase transition, lattice dynamics has not yet been investigated, which might contribute to the Fermi surface instability by itself. Optical phonons can be taken as probes of global structural instabilities as well as local electronic and charge modulations because of their intrinsic coupling to electronic and orbital subsystems.
Here, we describe a Raman scattering study of IrTe$_{2-x}$Se$_x$ ($x = 0$ and 0.45). Our major finding is the appearance of five zone-folded peaks for the 128 cm$^{-1}$ in-plane mode and six zone-folded peaks for the 165 cm$^{-1}$ out-of-plane mode below $T_C$, which is common to the pure and the Se-doped compound. This confirms the 1/5 charge modulation and further suggests that the ground state of IrTe$_2$ is in proximity to a 1/6 commensurate charge modulated phase. In addition, the temperature and Se-doping dependences of phonon modes point toward a significant role of Te 5$p$ orbitals in relation to the structural instability.

2. Experimental details

Single crystals of IrTe$_{2-x}$Se$_x$ ($x = 0$ and 0.45) were grown using a self-flux method described in [14]. The structural and transport properties of the investigated crystals were intensively characterized by resistivity and STM [22]. Our crystals showed the second phase transition at $T_S$, which was absent in other polycrystalline samples or low-quality crystals. As detailed in the supplemental material of [22], this is ascribed to a tiny inhomogeneous strain, leading to a suppression of the second phase transition.

Raman scattering experiments have been performed in a quasi-backscattering geometry with the excitation line $\lambda = 532$ nm of a solid-state laser ($P = 5$ mW). The laser beam was focused to a 100-μm-diameter spot on the surface of the crystal. Measurements were carried out in an evacuated closed-cycle cryostat with the temperature range of $T = 7$ to 288 K as well as on a heating stage with the temperature range of $T = 288$ to 530 K. The spectra were recorded using a DILOR-XY triple spectrometer equipped with a nitrogen-cooled charge-coupled device detector. A Ne lamp was used to calibrate the spectral position of a spectrometer. Samples were cleaved to obtain fresh surfaces before mounting them for the experiments. The cleaved surfaces were parallel to the $ab$ plane of the high-temperature trigonal phase.

To avoid any thermal history effects, Raman measurements were taken in one thermal cycle; Raman spectra were recorded while heating the samples after an initial cooling down to $T = 7$ K. Furthermore, we checked a couple of crystal pieces to examine a variation of structural and electronic properties with the samples. We found the essentially same phonon spectra for all studied samples.

3. Results and discussion

3.1. IrTe$_2$

Figure 1 compares the polarized Raman spectra of IrTe$_2$ between $T = 7$ and 288 K measured in $(xx)$ polarization. In the high-$T$ trigonal phase, we observe two sharp peaks at 128 and 165 cm$^{-1}$. The observed two phonon modes are in accordance with the factor group prediction for the space group $P3\bar{3}m1$:

$$\Gamma_{\text{Raman}}^{\text{HT}} = A_{1g}(xx, yy, zz) + E_g(xx, yy, xy, xz, yz).$$

The peak at 128 cm$^{-1}$ is assigned to the $E_g$ mode, which corresponds to in-plane stretching vibrations of Te atoms [24]. The peak at 165 cm$^{-1}$ with $A_{1g}$ symmetry involves out-of-plane motions of Te atoms, as depicted in figure 2. These assignments are further confirmed by calculated phonon dispersions (see the $\Gamma$ point of figure 6 in [18]): the doubly degenerate $E_g$
As the temperature is lowered below $T_C$, each mode splits into many sharp peaks, allowing for a fit of the phonons to individual Lorentzian profiles, as shown in the inset of figure 1(b). We are able to identify five zone-folded peaks for the 128 cm$^{-1}$ mode and six zone-folded peaks for the 165 cm$^{-1}$ mode, pointing to a quintupling and sextupling of the unit cell, respectively. The quintupling observed in the 128 cm$^{-1}$ mode is consistent with the 1/5 charge modulation. As to the sextupling, in the following section we will discuss its implications while comparing the results with IrTe$_{1.55}$Se$_{0.45}$. In addition to the strong sharp peaks, an additional peak around 81 cm$^{-1}$ is discernible. Consequently, the low-$T$ Raman spectra consist of three principal phonons in addition to zone-folded phonons.

In the low-$T$ phase, IrTe$_2$ possesses an average monoclinic $C2/m$ structure where the Ir–Ir bonds shrink uniformly along the monoclinic $b$-axis [11]. Ensuing works refined the true structure to a triclinic $P\overline{1}$ with modulated Ir–Ir bond lengths [18, 20, 23]. For the $C2/m$ space group the total irreducible representation for Raman-active modes is given as

$$\Gamma_{\text{Raman}}^{LT} = 2A_g(xx, yy, zz, xz) + B_g(xy, yz).$$
For the $P\bar{1}$ space group the factor group analysis yields a total of 21 Raman-active modes:

$$\Gamma_{Raman}^{LT} = 21A_g(xx, yy, zz, xy, xz, yz).$$

The comparison between the group theory and experimental data suggests that the principal modes are described by the average monoclinic symmetry while the folded modes reflect local lattice and bond modulations. This observation is related to the characteristics of Raman spectroscopy, which is more sensitive to local lattice distortions than a global symmetry change [26].

According to the correlation diagram of the symmetry transformation to the monoclinic phase, the $A_{1g}$ mode of the high-$T$ phase is correlated with the $A_g$ mode of the low-$T$ phase, and the double degenerate $E_g$ mode with the $(A_g + B_g)$ modes (see figure 2). Thus, the low-frequency peak at 81 cm$^{-1}$ is assigned to the symmetry-forbidden $B_g$ mode. Its weak intensity is due to a leakage of a selection rule in the $(xx)$ scattering configuration. The remaining phonons have $A_g$ symmetry. Their normal-mode displacements are sketched in figure 2 [24, 25]. In the following, we will examine the evolution of $E_g \rightarrow A_g$ and $A_{1g} \rightarrow A_g$ modes through $T_C$.

We now focus on the background signal. Unlike CDW or charge-orbital ordered systems, we cannot observe a depletion of the electronic background [27]. Rather, the featureless background is visible in the whole temperature range, which levels off upon cooling through $T_C$ (see the shaded area in figure 1). This is consistent with no opening of a gap at the Fermi surface [15–17].

In simple metals light scattering by electrons is normally not observable at low energy, because electrons will be collectively excited with the plasma frequency of several electron volts. In contrast, correlated electron systems show distinct electronic excitations and collective

**Figure 2.** The top panel depicts the eigenvectors of the 128 cm$^{-1}$ $E_g$ and the 165 cm$^{-1}$ $A_{1g}$ modes of the high-temperature trigonal phase. The bottom panel depicts the eigenvectors of the 81 cm$^{-1}$ $B_g$, the 128 cm$^{-1}$ $A_g$, and the 165 cm$^{-1}$ $A_g$ modes of the low-temperature monoclinic phase. The green (blue) balls represent Ir (Te) ions.
modes in the optical phonon energy range [28]. The observed featureless background indicates that electronic correlations are weak, being in line with the theoretical calculations that show negligible dynamical mean field theory corrections to density functional theory [20, 23]. However, the level-off of the background upon cooling through $T_C$ suggests its sensitivity to the structural phase transition, which leads to a change of local bonds and a partial depletion of density of states at $E_F$. Indeed, below $T_S$ the density of states is reconstructed to shift spectral weight downward from the region around 1 eV to the region around 0 eV [18].

To evaluate the electronic background, we integrate its signal over a frequency interval of 18–700 cm$^{-1}$. The temperature dependence of this integrated intensity is plotted in the inset (a) of figure 1. The intensity decreases drastically as the temperature is lowered through $T_C$. This is correlated with the site-dependent decrease of the local density of states at $E_F$ through $T_C$ [20, 23]. Thus, the decrease of the electronic background is ascribed to the suppression of light scattering by itinerant carriers. It is worth noting that the phonon modes retain a typical Lorentz shape with no hint of a Fano line shape. This suggests that the electron–phonon coupling is small.

![Figure 3](image.png)

**Figure 3.** Temperature dependence of frequencies, full widths at half maximum, and normalized integrated intensities of the 128 and 165 cm$^{-1}$ modes. The phonon intensities are corrected for the Bose thermal factor. The red lines are fits of the intensities to $I(T) \sim \exp(-k_BT/\Delta)$ with $\Delta = 51.3 \pm 0.6$ K (42.2 $\pm$ 0.5 K) for the 128 (165) cm$^{-1}$ mode. The dashed lines are a guide to the eye, and the vertical gray bars indicate the phase transition temperatures.

We next inspect the temperature dependence of phonon parameters measured upon heating after an initial cooling down to 7 K. The phonon peaks are fitted to a sum of Lorentzian profiles. The resulting frequencies, full widths at half maximum, and normalized integrated intensities for the 128 and 165 cm$^{-1}$ modes are summarized in figure 3 as a function of temperature. The errors are a symbol size. The parameters evolve in an intriguing way with an abrupt change at $T_C$, demonstrating the first-order character of the phase transition. Since our crystals are known
to have the second transition at \( T_2 \approx 180 \text{ K} \) [22], we examine a possible anomaly at the respective temperature. Only a subtle change is observed in the phonon parameters through \( T_2 \) (see the dashed lines in figure 3). The absence of apparent phonon anomalies suggests a delicate nature of the second transition, which is related to the appearance of new modulations with longer wavelengths [15, 22].

Below \( T_C \), the frequencies harden by 1–2 cm\(^{-1}\) and the phonon linewidths barely change. Their \( T\)-dependence is not ascribed to anharmonicities. Rather, a competing mechanism should be invoked, which is based on scattering of phonons by electrons. Noticeably, the integrated intensities increase exponentially as \( T \rightarrow 0 \). The intensity is calculated by integrating all zone-folded peaks. We can extract the empirical parameter \( \Delta = 51.3 \pm 0.6 \text{ K} (42.2 \pm 0.5 \text{ K}) \) from the 128 (165) cm\(^{-1}\) mode using the relation \( I(T) \sim \exp (-k_B T/\Delta) \). We recall that the Raman intensity of a phonon mode is determined by a change of the electronic polarizability tensor. The total electronic polarizability is approximated by the sum of Te–Te bond polarizabilities. In this regard, the strong variation of the phonon intensity is ascribed to a change of bond modulations and indicates a local bonding instability associated with the Te 5\( p \) states. It is possible that, the extracted empirical constant \( \Delta \) gives an energy scale for the rearrangement of the local bonding via structural distortions [15, 18].

Figures 4(a) and (b) detail the Raman spectra in the vicinity of \( T_C \) measured during heating and cooling. With increasing temperature through \( T_C^{heating} \approx 281 \text{ K} \), the zone-folded modes totally disappear. Upon cooling through \( T_C^{cooling} \approx 277 \text{ K} \), the zone-folded modes suddenly show up. In figures 4(c) and (d) we plot their frequency and integrated intensity in the warming and cooling processes. They exhibit a thermal hysteresis with width \( \Delta T = 4 \text{ K} \), which is in line with the hysteresis behaviors of lattice parameters, magnetic susceptibility, and resistivity [14, 15]. Through \( T_C \), the \( A_{1g} \) mode softens by 1 cm\(^{-1}\) while the \( E_g \) mode hardens by 2.5 cm\(^{-1}\). We note that a similar frequency shift has been reported in 1 \( T \)-TaS\(_2\) [29]. On a qualitative level, the opposite frequency shift between the in-plane and out-of-plane modes is explained by an increase of the ratio of interlayer to intralayer Te–Te distances as well as by an enhancement of the ionic character of the covalent Te–Te bonds. This leads to a change of the respective force constants. Upon cooling across \( T_C \), the intensity \( I(T) \) drops abruptly. The decrease of \( I(T) \) becomes more pronounced for the out-of-plane \( A_{1g} \) mode than the in-plane \( E_g \) mode. The softening and the strong suppression of the out-of-plane mode below \( T_C \) demonstrate that the weakening of interlayer Te–Te bonds plays a significant role in the structural transition. It is worth noting that the essentially same conclusion is drawn by first-principle calculations [18].

A close inspection of the phonon spectra reveals that they are slightly different between the cooling and heating processes. For the \( E_g \) mode, the zone-folded (Z) peak becomes more intense than the principal (P) peak across \( T_C \) upon cooling, which is opposite in the warming process [marked by the vertical arrows in figures 4(a) and (b)]. In contrast, we find no sizable effect in the \( A_{1g} \) mode. The selective sensitivity of the in-plane mode to the thermal history is evident from the ratio \( I_Z/I_P \) of the Z and P peak intensity as plotted in figures 4(e) and (f). Considering that the in-plane mode is closely tied to the intralayer bond and charge modulations, the stronger intensity of the Z mode relative to the P mode in the cooling process corroborates the robustness of the 1/5 modulation. On the contrary, upon warming it is destabilized while coexisting with multiple fractional modulations.
3.2. IrTe$_{2-x}$Se$_x$ ($x = 0.45$)

Now we will examine Se doping effects on the structural transition. Figure 5 shows the temperature dependence of the polarized Raman spectra for IrTe$_{1.55}$Se$_{0.45}$ measured upon heating. In the high-$T$ phase the phonon spectra consist of the intense principal $E_g$ and $A_{1g}$ modes with weak side peaks [see the fit in figure 5(b)]. The latter feature is totally absent in the pure IrTe$_2$ and points toward the presence of a precursor phase. Compared to IrTe$_2$, the frequencies shift slightly to higher energies by 1–2 cm$^{-1}$. This signals a stronger force constant of the Te–Te(Se) bonds because Se has a smaller ionic radius and is more electronegative than Te.

For temperatures below $T_C$, the two $A_g$ modes develop fine structures arising from the superstructure. In contrast to IrTe$_2$, the zone-folded peaks are not separable from each other due to disorder. However, we are able to identify a number of them as shown in figure 5(c).
Noticeably, the linewidth becomes much broader for the out-of-plane mode than for the in-plane mode. This is not the case for pure IrTe$_2$ [compare the inset of figure 1(b) with figure 5(c)]. The anisotropic effect of the Se substitution on the two phonons relies on the fact that the disparate Te–Se bonds break partly the covalent interlayer Te–Te bonds. A random distribution of the Te–Se bonds located in the Te–Te bonds leads to a spread-out of the out-of-plane phonon. However, the in-plane mode remains intact because of the secondary role of the Te–Te(Se) dimers for the CDW transition. That is, the dimerization of Ir atoms leads the formation of the Te–Te(Se) dimers in the plane. Our results give further support for the assertion that the destabilization of the interlayer Te–Te bonds is a prerequisite for the structural transition [14].

We fit the $T = 7$ K phonon spectra to a sum of Lorentzian profiles. We can clearly resolve six peaks for the 167 cm$^{-1}$ mode, whereas five peaks suffice to describe the 128 cm$^{-1}$ mode [see figure 5(c)]. Remarkably, we observe the same zone-folded feature as in the case of $x = 0$: a coexistence of the 1/5 and 1/6 lattice modulation. This seems to be incompatible with the transformation of the superstructure from 1/5(10$\bar{1}$) to 1/6(10$\bar{1}$) with Se doping. The invariant Raman result should not be ascribed to the insensitivity of optical phonons to the variations of the superstructure. Rather, it contains profound implications for the intrinsic ground state. For $x = 0$, a STM study [22] shows the instability of the $2a^*$ soliton lattice toward the $3a^*$ stripes in the 1/5(10$\bar{1}$) modulation. This leads to a coexistence of the 1/5 modulation with multiple modulations of longer wavelengths. In this case, the multiple modulations probed by optical phonons may correspond effectively to a 1/6 superstructure. Consequently, the phonon spectra

Figure 5. (a) Temperature dependence of polarized Raman spectra of IrTe$_{1.55}$Se$_{0.45}$ measured upon heating in (xx) polarization. (b) and (c) Representative fits of the phonon spectra to Lorentzian profiles at $T = 526$ and 7 K. $P_i$ ($i = 1$–5) denote five zone-folded phonons of the 128 cm$^{-1}$ mode and $Q_i$ ($i = 1$–6) denote six zone-folded phonons of the 167 cm$^{-1}$ mode.
have a concomitant signature for the 1/5 and 1/6 lattice modulation. This suggests a proximity of IrTe$_2$ to a commensurate 1/6 charge modulation.

Before proceeding, we would like to mention another interpretation. Since Raman spectroscopy is more bulk sensitive than STM, the multiple modulations seen by STM is related to a surface effect. In a bulk limit, the 1/6 modulation is stabilized while coexisting with the 1/5 charge modulation. Further studies are needed to elucidate this scenario.

In figure 6(a) we summarize the temperature dependence of the frequencies, full width at half maximum, and normalized integrated intensity of the 128 and 167 cm$^{-1}$ modes. The solid lines of the top panel are fits to the anharmonic model, $\omega_{ph}(T) = \omega_0 + A[1 + 2(e^{\hbar \omega_0/2k_BT} - 1)]$. The phonon intensity is corrected for the Bose thermal factor. The solid lines of the bottom panel are fits to $I(T) \sim \exp(-k_BT/\Delta)$ with $\Delta = 68.9 \pm 0.4$ K (74.3 $\pm$ 0.7 K) for the 128 (167) cm$^{-1}$ mode. The dashed lines are a guide to the eye. Hysteresis loops of frequency (b) and integrated intensity (c) of the 128 and 167 cm$^{-1}$ modes. The arrows indicate the direction of temperature sweep.

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In figure 6(a) we summarize the temperature dependence of the frequencies, full widths at half maximum, and normalized integrated intensities of the 128 and 167 cm$^{-1}$ modes. A discontinuous change of all phonon parameters is clearly observed at $T_C = 491$ K, confirming again the first-order structural transition. In addition, we examine the appearance of anomaly around $T_S = 284$ K. Unlike $x = 0$, we find some indication of the anomaly as seen from the discernible change of the linewidth through $T_S$ [see the dashed lines in the middle panel of figure 6(a)]. As the temperature is lowered through $T_C$, the frequencies of the two modes increase by 4–5 cm$^{-1}$ after a sudden jump by 7–8 cm$^{-1}$. Below $T_S$, the hardening is well described by anharmonic processes [see the solid lines in figure 6(a)]. This means that the low-$T$ phase is more stable in $x = 0.45$ than in $x = 0$. In figure 6(b) we zoom into the hysteresis around $T_C$. The hysteresis width $\Delta T = 25$ K and the frequency shift are much bigger than that for the case of $x = 0$. This is in line with the large increase of $T_C$ upon Se doping. As shown in figure 6(c), the drop of the intensity through $T_C$ is comparable to that of $x = 0$. Below $T_S$, the phonon intensity shows an exponential increase with $\Delta = 68.9 \pm 0.4$ K (74.3 $\pm$ 0.7 K) for the
in-plane (out-of-plane) mode. The extracted parameter becomes larger for $x = 0.45$ than for $x = 0$, implying that the rearrangement of the local bonding involving the Te(Se) $5p$ states becomes stronger upon Se doping.

Finally, we mention several contrasting phonon behaviors between $x = 0.45$ and $x = 0$: (i) a hardening of the $A_{1g}$ mode in $x = 0.45$ versus its softening in $x = 0$ and (ii) a sudden jump of width at $T_C$ in $x = 0.45$ versus its drop in $x = 0$ [compare the middle panel of figure 3 and figure 6(a)]. This highlights an intriguing effect of the Se doping on intralayer and interlayer bonds and electronic properties, which deserves further studies.

4. Conclusion

We have presented a Raman scattering study of the two dichalcogenides IrTe$_{2-x}$Se$_x$ ($x = 0$ and 0.45). Optical phonons have been used to probe the multifaceted aspects of a structural/electronic transition as functions of temperature and Se doping. The most remarkable feature is the observation of five zone-folded phonons for the 128 cm$^{-1}$ in-plane mode and six zone-folded phonons for the 165 cm$^{-1}$ out-of-plane mode, which is common to $x = 0$ and $x = 0.45$ in the low-temperature phase. This feature is taken as evidence for the presence of a 1/6 commensurate charge modulation in addition to a 1/5 modulation for both systems. Se doping primarily weakens interlayer Te–Te interactions while enhancing intralayer Te–Te(Se) bonds, leading to the strengthening of electronic and bond anisotropies. As a consequence, it enforces structural instabilities. However, there emerge intriguing features that are beyond such a scenario. Therefore, we call for further spectroscopic works to better understand the electronic and structural complexities caused by introducing disparate bonds into transition metal dichalcogenides.

Acknowledgements

We would like to thank V P Gnezdilov for fruitful discussions and assistance with Raman measurements. This work was supported by Korea NRF grants (no. 2012–046138) as well as by German-Israeli Foundation (GIF, 1171-486 189.14/2011), the NTH-School ‘Contacts in Nanosystems: Interactions, Control and Quantum Dynamics’, the Braunschweig International Graduate School of Metrology, and DFG-RTG 1953/1, Metrology for Complex Nanosystems. The work at Postech was supported by the Max Planck POSTECH/KOREA Research Initiative Program (grant no. 2011–0031558) through NRF of Korea funded by MEST. The work at Rutgers was supported by the National Science Foundation DMREF 1233349.

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