1. Introduction

During the last two decades, numerous studies have focused on two-dimensional (2D) nanomaterials because graphene sheets have been produced via mechanical exfoliation to obtain unusual physical and chemical properties. More recently, graphene-like 2D nanomaterials such as metal chalcogenides and phosphorene have attracted significant interest as they can be used to manufacture new or improved field-effect transistors, optoelectronic devices, and topological superconductors. Metal chalcogenides, include metal dichalcogenides and metal monochalcogenides, whose chemical formulae are MX and MX₂, respectively, where M represents a metal atom of molybdenum (Mo), tantalum (Ta), rhenium (Re), germanium (Ge) or tin (Sn); and X stands for atoms of either sulfur (S), selenium (Se), or tellurium (Te). MX has wrinkled honeycomb structures, similar to phosphorene, which shows an ultralow thermal conductivity. MX₂ has a hexagonal honeycomb structure with a high thermal conductivity. MX₂ can be used to manufacture new or improved 2D nanomaterials such as metal chalcogenides and phosphorene.

We systematically investigated the temperature-dependent bandgap energy and Raman shift on the bond length and bond energy, Debye temperature, and atomic cohesive energy for M(X, X₂) via bond relaxation methods. It is revealed that the thermal decay of both bandgap energy and phonon frequency arose from the thermal integration of the specific heat of Debye approximation. The results indicate that (i) the bandgap energy relaxation is due to the thermal excitation-induced weakening of the bond energy, and the phonon frequency was just a function of bond length and bond energy; (ii) the Debye temperature determines the nonlinear range at low temperatures; (iii) the reciprocal of the atomic cohesive energy governs the linear behavior at high temperatures. Thus, the outcomes of this study include fundamental information about photon, phonon, and the thermal properties of layered semiconductors, which are crucial to develop the new generations of thermal and electronic applications of devices based on layered semiconductors.

Bond-photon-phonon thermal relaxation in the M(X, X₂) (M = Mo, Re, Ta, Ge, Sn; X = S, Se, and Te)

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We systematically investigated the temperature-dependent bandgap energy and Raman shift on the bond length and bond energy, Debye temperature, and atomic cohesive energy for M(X, X₂) via bond relaxation methods. It is revealed that the thermal decay of both bandgap energy and phonon frequency arose from the thermal integration of the specific heat of Debye approximation. The results indicate that (i) the bandgap energy relaxation is due to the thermal excitation-induced weakening of the bond energy, and the phonon frequency was just a function of bond length and bond energy; (ii) the Debye temperature determines the nonlinear range at low temperatures; (iii) the reciprocal of the atomic cohesive energy governs the linear behavior at high temperatures. Thus, the outcomes of this study include fundamental information about photon, phonon, and the thermal properties of layered semiconductors, which are crucial to develop the new generations of thermal and electronic applications of devices based on layered semiconductors.
substantially degrade in highly integrated electronic components. Usually, the heat in 2D nanomaterials mainly propagates via acoustic phonons,\textsuperscript{27,28} which determine its thermal conductivity. Therefore, it is very important to study the temperature dependence of the lattice vibrations of metal chalcogenides as nanomaterials. Raman spectroscopy is a very good method for the \textit{in situ} analysis of both phonon scattering and lattice distortion in the 2D nanomaterials. Therefore, the thermal response of Raman spectroscopy of the 2D materials was measured in some studies. For monolayer ReSe\textsubscript{2} (ref. 29), the frequencies of all the observed phonon modes exhibited nonlinear temperature dependence with incident laser wavelengths of 532 nm at low temperatures. Moreover, the temperature-dependent Raman spectroscopy of the SnSe sample\textsuperscript{30} shows intrinsic non-harmonic phonon scattering. In general, the temperature dependence of the Raman active modes of the 2D nanomaterials was analyzed using the Grüneisen\textsuperscript{31-33} model, which indicates that the non-harmonic properties are due to the decay of an optical phonon into multiple acoustic phonons. However, the calculation of the nonlinear phonon properties at low temperatures using the Grüneisen model does not agree well with the observations.

In this study, we used a Bond-Order-Length-Strength (BOLS) approximation,\textsuperscript{35,36} which intrinsically probes bond–phonon thermal softening properties across a wide temperature range. A simulation of the temperature-dependent bandgap energy and Raman shift of these 2D films revealed valuable quantitative information, including the Debye temperature (\(\theta_D\)), atomic cohesive energy (\(E_z\)), and the referential frequency (\(\omega_{\text{ref}}\), \(L\) denotes \(A_{1g}, E_{1g}, A_{2g}\), and \(B_{1g}\) modes). We confirm that the Debye temperature determines the nonlinear random range at low temperatures, while the atomic cohesive energy determines the linear behavior at high temperatures.

2. Principles

2.1. Bond-phonon correlation

It is well-known that a Raman shift can be described using \(\omega = \omega(0) + \Delta \omega\). Here, \(\omega(0)\) is the Raman frequency of the reference point, and \(\Delta \omega\) is the perturbation of the temperature (\(T\)) with respect to the Raman frequency. \(\omega(0)\) varies with the frequency of the substrate material and the wavelength of the incident light. However, the trend of the perturbation \(\Delta \omega\) remains unchanged when thermal excitation is used. According to the interatomic potential function, \(u(r)\) can be expanded using a Taylor series around its equilibrium position, and the Raman shift can be derived as:\textsuperscript{37}

\[
\frac{1}{2} \Delta \omega^2 \approx \frac{1}{2} \frac{\partial^2 u(r)}{\partial d^2} \bigg|_{d = d_e} (r - d_e)^2 + o \left[ (r - d_e)^3 \right] \quad (1)
\]

The term \((n = 0)\) corresponds to the equilibrium single bond energy at 0 K; the term \((n = 1)\) is the force at equilibrium; the term \((n = 2)\) corresponds to the thermal vibration energy, which dominates the Raman shift; the term \((n \geq 3)\) correspond to the nonlinear vibrations. So, omitting the higher-order terms, one can obtain the following relationship:

\[
\frac{1}{2} \Delta \omega^2 \approx \frac{1}{2} \frac{\partial^2 u(r)}{\partial d^2} \bigg|_{d = d_e} x^2 \approx \frac{1}{2} \frac{E_z}{\mu_d^2} x^2 \quad (2)
\]

According to eqn (2), the Raman shift \(\omega\) can be described by using \(\Delta \omega\), which is a function of bond length \(d_e\) and bond energy \(E_z\) in the \(z\)th atomic layer for the entire Raman spectroscopy. The \(\mu = m_1m_2/(m_1 + m_2)\) is the reproduced mass of the representative bond.\textsuperscript{38} Thus, the Raman shift can be derived as: \(\omega(d_e, E_z) = (E_z/\mu)^{1/2}/d_e\).

2.2. Bond-phonon correction

The bandgap energy \(E_g\) can be calculated using the first-order Fourier coefficient of the crystal potential \(V_{\text{cr}}\) function. \(E_g = 2 |V_1| \times \langle E_z \rangle\), where the bandgap energy is proportional to the single bond cohesive energy \(E_z\), and \(V_1 = \int V_{\text{cr}}(r)e^{ik \cdot r} \, dr\). Here, \(e^{ik \cdot r}\) is the Bloch wave function for equilibrium conditions. This means that the crystal potential function determines the intrinsic bandgap energy \(E_g\). The expression of the bandgap energy for a given perturbation temperature can be written as:

\[
E_g(T) = E_g(0)(1 + \Delta_T) \quad (3)
\]

here, \(E_g(T)\) and \(E_g(0)\) represent the bandgap energies at the measured temperature and 0 K, respectively. \(\Delta_T \int_0^T \eta(t) \, dt\) is the perturbation of temperature with respect to the bandgap energy.

2.3. Bond length and bond energy thermal relaxation

The relationship between the bond length and bond energy for thermal relaxation can be described using the following equation:

\[
\begin{align*}
\frac{d(T)}{d_e} &= \frac{d_e}{E_z} \left(1 + \int_0^T \frac{\omega(t) \, dt}{E_z} \right) \\
E(T) &= E_z \left[1 - \frac{\int_0^T \eta(t) \, dt}{E_z} \right]
\end{align*} \quad (4)
\]

here, \(\alpha(t)\) is the thermal expansion coefficient; \(d_e\) and \(E_z\) denote the bond length and bond energy of the film materials without thermal stimuli, respectively. Eqn (4) indicates that the bond length expands, and the bond energy weakens upon stimulation via heating for the entire electron and Raman spectrum. The integration of the specific heat \(\eta(t)\) can be described by the following relationship:

\[
\int_0^T \eta(t) \, dt = \int_0^T \frac{C_v(T/\theta_D)}{z_i} \, dt = \frac{9RT}{E_z} \left( \frac{T}{\theta_D} \right)^3 \int_0^{\theta_D/T} x^{3} \, dx \quad (5)
\]

here, \(z_i\) is the atomic effective coordination number, \(\theta_D\) is the Debye temperature, \(E_z\) is the single bond energy, and \(x = \theta_D/T\) is the reduced form of the temperature. When the external temperature is greater than the Debye temperature, the specific heat \(C_v\) is approximately equal to a constant value of 3\(R\), where \(R\) represents the ideal gas constant. The atomic cohesive energy \(E_z\)
\[\omega(T) = \omega(0) + a(T)e^{-\Delta t/T}\]

where \(\omega(T)\) is the measured value for the Raman frequency, \(\Delta \omega(T) = \omega(T) - \omega(0)\) is the variation of the Raman frequency, \(\omega(1)\) denotes the vibration frequency of a dimer, which comes from the Raman frequency of the reference point \(\omega(0)\). Here, the reference frequency can vary with an incident laser wavelength, but the resulting shift is independent of the incident laser energy. Furthermore, \(d(T)\) and \(E(T)\) denote the bond length expansion and bond energy weakening with increase in temperature, respectively. Therefore, the Raman frequency and bandgap energy of these 2D films can be expressed as a function of bond length and bond energy for thermal excitation.

Combining eqn (1) to eqn (6), the thermal relaxation of the Raman frequency and bandgap energy can be expressed as:

\[
\begin{align*}
\frac{\Delta \omega(T)}{\Delta \omega(T_0)} & = \frac{d_\omega}{d_\omega(T_0)} \left[ E(T) \right]^{1/2} \\
\frac{E_g(T)}{E_g(T_0)} & = \exp \left( \int_0^T \frac{\eta(t)}{E_z} dt \right) \\
\end{align*}
\]

(6)

According to eqn (7), the atomic cohesive energy is the only adjustable parameter for the calculation of the temperature-dependent Raman shift and bandgap energy. Because the thermal expansion coefficient \(a(t)\) is a very small value, about \(10^{-6} \text{ K}^{-1}\), the following is valid: \(1 + \int_0^T a(t) dt \leq 1 + 0.05\).

Therefore, we can neglect the effects of thermal expansion on the temperature dependence of the Raman shift. Here, a proportional relationship was established to be studied between the temperature dependence of the Raman shift and bandgap energy because the exact values of the implied constants in the expression cannot reveal the relative variation in the respective physical quantities. The main objective of our study was to determine the bond contributions to electron and phonon spectroscopy and to extract the quantitative information from complex measurements carried out by other researchers.

3. Results and discussion

3.1. The vibrational modes of M-(X, X$_2$)

The interatomic force of M(X, X$_2$) is made up of covalent bonds and the van der Waals (VDW) forces. Strong covalent bonds provide the in-plane stability to 2D films, whereas relatively weak VDW forces are sufficient to keep the stack together. In Fig. 1, the dotted line denotes the weakening of the interlayer VDW force; the purple spheres are metal atoms; the green balls are group VIA atoms. Fig. 1(a) shows the two Raman active vibration modes of MX$_2$. \(E_{2g}\) is the in-plane vibration mode, which comes from the opposite direction of the vibration between two X atoms and intermediate M atoms. \(A_{1g}\) is the out-of-plane vibration mode, which is derived from the opposite direction of the vibration of two X atoms in a unit cell. Contrastingly, the MX films show out-of-plane modes (\(A_{2g}\)) and in-plane modes (\(B_{3g}\)) (Fig. 1(b)) because MX shows wrinkled honeycomb structures, which is different from the hexagonal honeycomb structure of MX$_2$. The vibration modes of M(X, X$_2$) can not only be used to identify various nanomaterials but also provide the necessary basis for the study of the temperature dependence of the Raman shift of these 2D films.

3.2. Thermal relaxation of Raman shifts

Fig. 2 and 3 depict the BOLS reproduction observation results of the temperature-dependent \(A_{1g}\) modes and \(E_{2g}\) modes of MX$_2$.
The trends of all $E_{1}^{2g}$ and $A_{1g}$ modes turn gradually from nonlinear to linear during heating because the specific heat is $\eta(t) \propto T^3$ at low temperatures, due to which the Raman shift varies slowly with temperatures. However, the specific heat $\eta(t) = 3R$ (R is the ideal gas constant) at high temperatures, so the Raman shift display a linear decrease with the increase in temperature. Thus, the bond relaxation theory confirms that the Debye temperature determines the nonlinear range of the Raman shift at low temperatures. By calculating the temperature dependence of the Raman shift of MX$_2$, the Debye temperature ($\theta_D$), the referential frequency $\omega_L(1)$ (L denote $A_{1g}$ and $E_{1}^{2g}$ modes), and atomic cohesive energy ($E_z$) can be listed in Table 1. These parameters are essential for calculating the temperature-dependent Raman shift of these 2D films.

Fig. 2  The BOLS reproduction of the measured Raman $A_{1g}$ modes for (a) few-layer$^{48,49}$ (FL) and monolayer$^{48}$ (ML) MoTe$_2$, (b) (Ta, Sn, Re)-Se$_2$,$^{50-54}$ and (c) (Sn, Ge)-(S$_2$, Se$_2$).$^{44,54}$

Fig. 3  Theoretical reproduction of the measured Raman $E_{1}^{2g}$ modes for (a) monolayer$^{52,49}$ (ML), bilayer$^{52,48}$ (BL), and few-layer$^{48,49}$ (FL) MoTe$_2$, (b) (Ta, Sn, Re)-Se$_2$.$^{50-55}$

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According to eqn (7), the physical mechanism of the temperature-dependent Raman shift originates from the thermal excitation-induced bond length expansion and bond energy weakening.

Fig. 4 shows the BOLS approximation calculated temperature-dependent $A_3$ and $B_{3g}$ modes of the MX films. The referential frequency $\omega_L(1)$ (L denote $A_3$ and $B_{3g}$ modes), Debye temperature $\theta_D$, and atomic cohesive energy $E_z$, which is hardly possible using the Varshni’s semiempirical relationship and the Grüneneisen model, is calculated in Table 1. From the table, it is clear that the Debye temperature for the SnS films was greater than for SnSe and GeSe, so the low-temperature nonlinear range of the former was distinctly larger than the latter. Thus, the results indicate that the Debye temperature determines the nonlinear range of the Raman shift of these 2D films at low temperatures.

### 3.3. $E_z$ resolved a linear behavior at high temperatures

We first studied the temperature-dependent Raman shift of the monolayer (ML), bilayer (BL), and few-layer (FL) of the MoTe$_2$ film, which is applicable to the BOLS approximation as well, see Fig. 2(a) and 3(a). The BOLS approximation demonstrates that the atoms under coordination shorten the bond length and strengthen the bond energy, which means that the $E_{2g}$ modes experience a blue shift, and the $A_{3g}$ modes undergo a redshift as the atomic layer number decreases. This was found to be similar to the layer number dependent Raman shift of MoS$_2$. As a result, when there was no thermal stimulus, the phonon frequency of the few-layers of the $A_{1g}$ modes of MoTe$_2$ was larger than that of the monolayer, but the phonon frequency of the $E_{1g}$ modes follows the opposite trend: $\omega_{ML} > \omega_{BL} > \omega_{FL}$.

In addition, since MoTe$_2$ with a different atomic layer number had the same atomic cohesive energy, it shows the same behavior at high temperatures. Therefore, the BOLS approximation can resolve not only the layer number effect of the Raman shift of MoTe$_2$ film but can also clarify its temperature dependence.

Moreover, the atomic cohesive energy $E_z$ determines the linear behavior at high temperatures of the temperature-dependent Raman shift for different nanomaterials in the same temperature range. When the temperature changes from 0 to 600 K, the atomic cohesive energy (see Table 1) shows the following relationships: $E_z$-TaSe$_2$ (5.23 eV) > $E_z$-SnSe$_2$ (4.45 eV) > $E_z$-MoTe$_2$ (4.22 eV) > $E_z$-ReSe$_2$ (3.39 eV). Compared to the other three materials, TaSe$_2$ exhibit the largest linear behavior at high temperatures, so the rate of decrease in the Raman shift is the fastest for continuous heating. In addition, as shown in Fig. 2(c), the behavior for SnS$_2$...
at high temperatures was clearly larger than that for GeSe$_2$ for temperatures between 0 and 300 K. So, the order of their atomic cohesive energy calculated using the bond relaxation theory was $E_c$-SnS$_2$ (3.51 eV) > $E_c$-GeSe$_2$ (2.29 eV). A similar situation occurs for the Raman shift temperature dependence of SnS, SnSe, and GeSe. The agreement between the BOLS calculations and the experimental measurements confirms that the atomic cohesive energy $E_c$ governs the behavior at high temperatures for thermal relaxation in these Raman spectroscopies.

3.4. Thermal relaxation of bandgap energy

The bandgap energy of the 2D nanomaterials is an important foundation for its applications in semiconductors and optoelectronic devices. Because the self-heating of the device is unavoidable, when an electronic equipment is used, this directly affects the bandgap energy of nanomaterials. This means it is important to understand the physical mechanism of the temperature-dependent of the bandgap energy of nanomaterials. According to eqn (7), the thermal excitation increases the bond energy, which causes the thermal softening of the bandgap energy of these 2D film materials (see Fig. 5). The Debye temperature determines the nonlinear range at low temperatures, while the atomic cohesive energy governs the linear behavior at high temperatures, which is similar to the temperature dependence of the Raman shift of these 2D films. The BOLS approximation derived both atomic cohesive energy $E_c$ and bulk bandgap energy $E_{0s}$, which were used to calculate the temperature dependence of the bandgap energy of these 2D films (see Table 1). Thus, these results not only show the reliability of the bond relaxation theory when the temperature dependence of bandgap energy was calculated but also confirm that the Debye temperature and the atomic cohesive energy play an important role in the thermal softening of the bandgap energy.

4. Conclusion

In summary, we have investigated in detail the temperature dependence of the Raman shift and bandgap energy for M(X, X$_2$) using the BOLS approximation. The thermal softening of the Raman shift was caused by bond length elongation and bond energy expansion; the photon energy originates from the thermal excitation-induced weakening of the bond energy. The good agreement between the theoretical predictions and the experimental observations confirms that the Debye temperature determines the nonlinear range at low temperatures, while the atomic cohesive energy governs the linear behavior at high temperatures.
temperatures for the thermal relaxation of the photon and phonon spectroscopy. Therefore, this study confirms the value of the bond relaxation theory, which enables the extraction of the quantitative information of bond length and bond energy from electron spectroscopy and phonon spectroscopy. The research method described in this study offers both new insight and effective tools for further exploring the thermal excitation behavior of other 2D films.

**Conflicts of interest**

There are no conflicts to declare.

**Acknowledgements**

This study was kindly supported by the National Natural Science Foundation of China (No.: 41425014), Key Laboratory Project of Jiangxi Province (No.: 201718CD40010) and Two 1000 Talents Plan Project of Jiangxi Province (No.: S2018CQKJ0755).

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