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SPECTRAL AND TEMPERATURE DYNAMICS OF PHOTON ABSORPTION IN MONATOMIC TRANSITION METAL DICHALCOGENIDES

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This study delves into the interband absorption dynamics of polarized radiation in monatomic layers of transition metal dichalcogenides (TMDs), a field critical to advancing nano- and optoelectronics. Using theoretical modeling and computational analysis, the spectral and temperature dependencies of one- and two-photon absorption coefficients were examined for linearly and circularly polarized light. Results reveal a pronounced increase in absorption coefficients with frequency, reaching a peak before declining, a behavior modulated by the material's bandgap and temperature. The analysis further highlights the substantial impact of light polarization on absorption characteristics, with temperature-dependent shifts in the maximum absorption frequency. These findings contribute to a deeper understanding of the optical properties of 2D TMD materials and pave the way for their application in designing efficient photodetectors, light-emitting diodes, and other next-generation optoelectronic devices.

Keywords: *Monoatomic layers; Transition metal dichalcogenides; Photon absorption; Spectral dependency; Temperature influence; Polarization effects*
PACS: 78.67.-n, 78.20.Bh, 42.65.-k

INTRODUCTION

Although transition metal dichalcogenides have been studied for more than 50 years [1–3], scientific interest in two-dimensional (2D) structures, in particular monoatomic layers of transition metal dichalcogenides (TMDs), has increased even more since the creation of graphene [4, 5]. Monatomic layers of TMD represent weakly (van der Waals) and strongly (covalently) bonded atoms, which makes it possible to isolate from a sample a 2D layer with a thickness equal to the size of one elementary lattice. The development of such technologies and recent advances in the production of new types of devices based on them [6] have opened up new possibilities for the use of 2D TMD layers in the field of nanoelectronics and optoelectronics [7].

One of the unique properties of TMDs such as MoS2, MoSe2, WS2 and WSe2 is that when two-dimensional monoatomic layers are produced from a bulk sample, they become a direct gap material as the number of layers is reduced. In particular, monoatomic single-layer transition metal dichalcogenides have the nature of direct-gap semiconductors [8]. Monoatomic layers of such TMDs are today widely used in the field of nano-, optoelectronics and photonics [9-12].

It should be noted that the excitonic optical properties of monoatomic layers of TMD have been widely studied in both experimental and theoretical aspects (see, for example, [13-19] and references therein). However, single- and multiphoton absorption of interband or single-band polarized light in semiconductors and its linear circular dichroism [20–23] have been theoretically poorly studied. Therefore, in this work, the frequency and temperature dependence of the interband single- and multiphoton absorption coefficient of linearly and circularly polarized light in monatomic TMD layers was studied.

METHODS

The investigation of interband photon absorption in monatomic layers of transition metal dichalcogenides (TMDs) was carried out using a combination of theoretical modeling and computational analysis. The study focused on the spectral and temperature-dependent behavior of one- and two-photon absorption coefficients under polarized radiation.

The optical transitions were analyzed using models designed for semiconductors with diamond and zinc blende lattice structures, adapted for the unique properties of 2D monatomic layers. The single-photon absorption coefficient was derived using the equilibrium distribution function of charge carriers and the composite matrix element for interband transitions. For two-photon absorption, the squared modulus of the composite matrix element was incorporated alongside the law of energy conservation for multiphoton transitions.

The temperature dependency of the bandgap energy was modeled using the Varshni and Passler formulas, which account for thermal effects specific to the materials studied. Effective masses of electrons and holes were treated as functions of temperature to reflect their sensitivity to thermal variations. The Hamiltonian was formulated to include terms representing the mixing of light and heavy hole states with conduction band states. This formulation ensured accurate incorporation of spin-orbit coupling effects, particularly relevant for narrow-gap TMD semiconductors.

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Polarization effects were considered by defining the polarization vector relative to the monatomic layer plane. Coherent saturation effects were analyzed using the Rabi parameter to evaluate the nonlinear optical response under varying light intensities. Calculations were conducted using the computational software "Maple," leveraging constants and material-specific parameters sourced from established databases and prior studies to ensure precision and reliability.

The absorption coefficients were analyzed for trends and validated by comparing the results with existing theoretical and experimental data. Spectral and temperature dependencies were visualized through graphs to illustrate key trends, such as peak absorption frequencies and their shifts with temperature and bandgap width. This methodological framework provided a comprehensive understanding of the optical properties of monatomic TMD layers and their behavior under polarized radiation.

SPECTRAL AND TEMPERATURE DEPENDENCES OF THE COEFFICIENT OF ONE- AND TWO-PHOTON LIGHT ABSORPTION

The spectral and temperature dependences of the coefficient of interband single-photon light absorption $\left(K_{c;\mathrm{V}}^{(1)}\right)$ in a monatomic TMD layer are determined by the relation [20, 25]

$$K_{c;V}^{(1)}=\frac{2\pi}{\hbar}\frac{\hbar\omega}{I}\sum_{\vec{k}_\perp}\left\langle\left|M_{c;V}^{(1)}(\vec{k})\right|^2\right\rangle\cdot\left[f_c(\vec{k}_\perp)-f_V(\vec{k}_\perp)\right]\delta\left(E_c(\vec{k}_\perp)-E_V(\vec{k}_\perp)-\hbar\omega\right) \tag{1}$$

where $M_{c;\mathrm{V}}^{(1)}(\vec{k})$- is the composite matrix element of the interband optical transition, $f_c(\vec{k}_\perp)\left(f_V(\vec{k}_\perp)\right)$- is the distribution function of electrons (holes) in the conduction band (valence band), $\omega(I)$-is the frequency (intensity) of light, $E_c(\vec{k}_\perp)=\left(\tilde{E}_g^2+\gamma^2k_\perp^2\right)^{1/2}$, $\left(E_V(\vec{k}_\perp)=-\left(\tilde{E}_g^2+\gamma^2k_\perp^2\right)^{1/2}\right)$- is the energy dispersion of electrons (holes), $\vec{k}_\perp$ - is the two-dimensional wave vector of current carriers , directed in the plane of the TMD layer, $\gamma=\hbar\left(\tilde{E}_g/m^*\right)^{1/2}$, $\tilde{E}_g=E_g/2$, E_g- is the bandgap width, m^* - is the effective mass of current carriers, the sign $\langle\ldots\rangle$ means angular averaging over the solid angles of the two-dimensional wave vector, the rest are well-known quantities.

Following [24] and taking into account the law of conservation of energy, it is not difficult to obtain that the square of the modulus of the composite matrix element of a single-photon optical transition we obtain:

$$\left|M_{c;V}^{(1)}(k_\omega)\right|^2=\left(\frac{eA_0}{c\hbar}\right)^2\frac{E_g^2+2\gamma^2k_\omega^2}{E_g^2+4\gamma^2k_\omega^2}p_0^2\left[e'^2_\perp-2\frac{\gamma^2k_\omega^2}{E_g^2+2\gamma^2k_\omega^2}_{x}\left(e'^2_x-e'^2_y\right)\right], \tag{2}$$

where $\overrightarrow{e'}_\perp=\left(e'_x,e'_y\right)$ - is the two-dimensional light polarization vector directed along the plane of the layer, $k_\omega=\sqrt{(\hbar\tilde{\omega})^2-\tilde{E}_g^2}/\gamma$ - is the wave vector of current carriers participating in a single-photon optical transition and is determined using the δ- function in (1). Then $K_{c;\mathrm{V}}^{(1)}$ becomes

$$K_{c;V}^{(1)}=\frac{\pi^2e^2}{\hbar n_\omega c}\frac{p_0^2\gamma n^{(2)}}{m_0\tilde{E}_g^2}\frac{\left(\xi_1^2-1\right)^{1/2}}{\xi_1}\cdot e^{E_g(1-\xi_1)/(2k_BT)}\left\langle\frac{7+\xi_1^2}{3+\xi_1^2}\left\langle\frac{1}{2\pi}\left[e'^2_\perp+\frac{1-\xi_1^2}{7+\xi_1^2}\left(e'^2_x-e'^2_y\right)\right]\right\rangle\right\rangle, \tag{3}$$

here $e^{E_F/k_BT}=\pi\gamma^2n^{(2)}/\left[k_BT\left(\tilde{E}_g+k_BT\right)\right]^{-1}e^{\frac{\tilde{E}_g}{k_BT}}$, $\xi_1=\hbar\omega/E_g$, with the help of which the Fermi energy is determined depending on temperature, two-dimensional concentration $n^{(2)}$ and band parameters[1].

Figure 1 shows the spectral and temperature relationships of the inter-zone one-photon absorption coefficient $K_{c;\mathrm{V}}^{(1)}(\omega,T)$ of light in monoatomic layers of metal dichalcogenides: the 1st line in the picture corresponds to $E_g=0.3\ eV$, the 2nd line corresponds to $E_g=0.5\ eV$ quantities and $500\ K<T<600\ K$ to the temperature range. $E_g=0.3\ eV$ and 200 K of $K_{c;\mathrm{V}}^{(1)}(\omega,T)$ were calculated. It can be seen from this figure that the spectral and temperature coupling of $K_{c;\mathrm{V}}^{(1)}(\omega,T)$ increases with the width of the forbidden zone, and as the temperature decreases, the one-photon absorption $K_{c;\mathrm{V}}^{(1)}(\omega,T)$ coefficient of light in monoatomic layers of metal dichalcogenides increases significantly: in particular, the value of $K_{c;\mathrm{V}}^{(1)}(\omega,T=200\ K)$ calculated for the $E_g=0.3\ eV$ forbidden zone is greater than the value calculated for the $E_g=1\ eV$ forbidden zone will be several thousand times larger. This is because e^{E_F/k_BT} takes large values in small temperature fields and large bandgap values (such temperature coupling is suitable for 2D current carrier systems).

In particular, in monoatomic layers of dichalcogenides of transition metals with $E_g=0.3\ eV$ forbidden zone, the magnitude of e^{E_F/k_BT} decreases 10 times when the temperature increases from 200 K to 270 K. For this reason, we recorded the spectral and temperature relationship of $K_{c;\mathrm{V}}^{(1)}(\omega,T)$ with respect to the field of $500\ K<T<600\ K$

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[1] In what follows we use the following notations for the N photon optical transition: $\xi_N=N\xi_1$ and $k_{N\omega}=\left[(N\hbar\omega)^2-E_g^2\right]^{1/2}/(2\gamma)=E_g\left[N^2\xi_1^2-1\right]^{1/2}/(2\gamma)$.

temperatures in Fig. 2 b). It can be seen from this figure that at a certain temperature value, the magnitude of $K_{c;\mathrm{V}}^{(1)}(\omega,T)$ increases with increasing frequency, reaches a maximum, and then decreases. However, at a specific value of $\hbar\omega$, $K_{c;\mathrm{V}}^{(1)}(\omega,T)$ decreases with increasing temperature, and this nature of temperature dependence is explained by temperature dependence of magnitude $\varsigma(T)=4\,e^{E_F/k_BT}/\left(\gamma^2\pi n^{(2)}E_g\right)$.

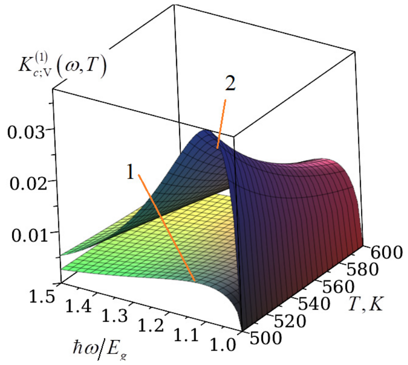
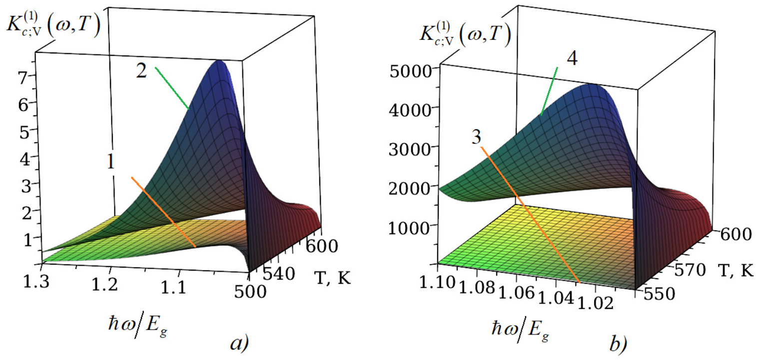


Figure 1. Spectral and temperature correlations of interband one-photon absorption coefficient $K_{c;\mathrm{V}}^{(1)}(\omega,T)$ of linearly polarized light in monoatomic layers of transition metal dichalcogenides: 1st line $E_g=0.3\ eV$, 2nd line $E_g=0.5\ eV$ in the figure correspond to quantities. In the calculations, the calculated maximum value of $K_{c;\mathrm{V}}^{(1)}(\omega,T=200\ K)$ for $E_g=0.3\ eV$ was taken as one unit.



a) b)

Figure 2. Spectral and temperature correlations of interband one-photon absorption coefficient $K_{c;\mathrm{V}}^{(1)}(\omega,T)$ of linearly polarized light in monoatomic layers of transition metal dichalcogenides: a) in the figure, line 1 corresponds to the quantity $E_g=1.0\ eV$, line 2 corresponds to the quantity $E_g=1.2\ eV$, b) line 3 corresponds to the quantity $E_g=1.5\ eV$, line 4 corresponds to the quantity $E_g=2.0\ eV$ will come In the calculations, the maximum value of $K_{c;\mathrm{V}}^{(1)}(E_g=1\ eV,\omega,T)$ was taken as a unit.

The $K_{c;\mathrm{V}}^{(1)}(\omega,T)$ bond reaches a maximum of $K_{c;\mathrm{V}}^{(1,\max)}(\omega_{\max},T)$ at frequencies $\omega_{\max}\approx 1.072E_g/\hbar$ and $\omega_{\max}\approx 1.125E_g/\hbar$ at temperatures $T=300\ K$ and $T=600\ K$, respectively, that is, $K_{c;\mathrm{V}}^{(1,\max)}(\omega_{\max},T)$ increases with increasing temperature (see Table 1).

Table 1. This table presents the mathematical relationship between the band gap energy E_g of semiconductor materials, the temperature T, and the peak absorption frequency $\omega_{\max}$ in the optical domain

	$T=300\ K$	$T=600\ K$
$E_g=0.3\ eV$	$\omega_{\max}\approx 1.072E_g/\hbar$	$\omega_{\max}\approx 1.125E_g/\hbar$
$E_g=0.5\ eV$	$\omega_{\max}\approx 1.058E_g/\hbar$	$\omega_{\max}\approx 1.10E_g/\hbar$
$E_g=1\ eV$	$\omega_{\max}\approx 1.03E_g/\hbar$	$\omega_{\max}\approx 1.058E_g/\hbar$

In the calculations, the calculated maximum value of $K_{c;\mathrm{V}}^{(1)}(\omega,T=500\ K)$ for $E_g=1.0\ eV$ was taken as one unit. $K_{c;\mathrm{V}}^{(1)}(\omega,T)$ bonds $T=550\ K$, $T=600\ K$ and $T=700\ K$ correspondingly at temperatures $K_{c;\mathrm{V}}^{(1)}(E_g=1\ eV,\omega,T)$ reach the maximum value of magnitude $\omega_{\max}\approx 1.043E_g/\hbar$, $\omega_{\max}\approx 1.046E_g/\hbar$ and $\omega_{\max}\approx 1.053E_g/\hbar$ frequencies, $K_{c;\mathrm{V}}^{(1)}(E_g=1.2\ eV,\omega,T)$ reach the maximum value of magnitude $\omega_{\max}\approx 1.036E_g/\hbar$,$\omega_{\max}\approx 1.041E_g/\hbar$ and $\omega_{\max}\approx 1.045E_g/\hbar$ frequencies, $K_{c;\mathrm{V}}^{(1)}(E_g=1.5\ eV,\omega,T)$ reach the maximum value of magnitude $\omega_{\max}\approx 1.029E_g/\hbar$, $\omega_{\max}\approx 1.032E_g/\hbar$, and $\omega_{\max}\approx 1.037E_g/\hbar$ frequencies, $K_{c;\mathrm{V}}^{(1)}(E_g=2\ eV,\omega,T)$ reach the maximum value of magnitude $\omega_{\max}\approx 1.022E_g/\hbar$,$\omega_{\max}\approx 1.024E_g/\hbar$, and $\omega_{\max}\approx 1.02E_g/\hbar$ frequencies, i.e. with an increase in temperature, the frequency corresponding to the maximum values of the $K_{c;\mathrm{V}}^{(1,2,3,4)}(\omega,T)$ quantities increases somewhat, although not significantly, but this maximum frequency decreases with an increase in the width of the forbidden zone.

The coefficient of two-photon interband absorption of light is defined as

$$K_{c;V}^{(2)} = \frac{2\pi}{\hbar}\frac{2\hbar\omega}{I}\left\langle\left|M_{c;V}^{(2)}(\vec{k})\right|^2\right\rangle\cdot[f_c(k)-f_V(k)]\delta(E_c(k)-E_V(k)-2\hbar\omega), \quad (4)$$

Then, given the expression for the squared modulus of a composite matrix element:

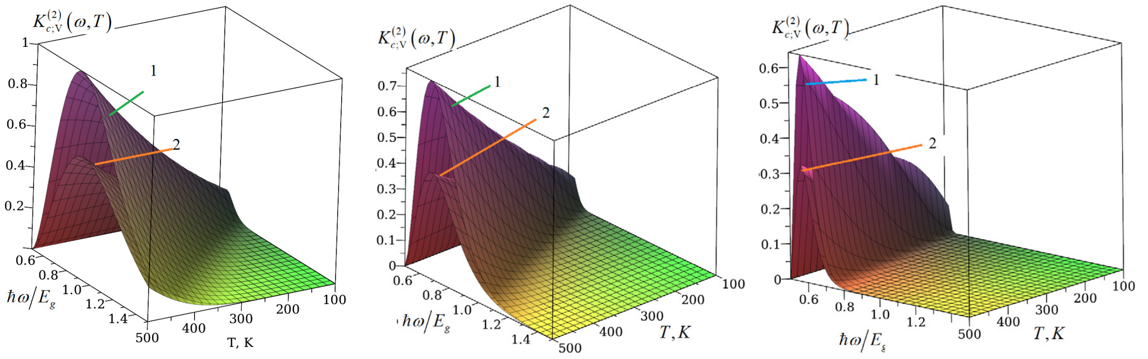
$$\left|\mathrm{M}_{cV}^{(2)}(\vec{k}_\perp,\vec{e})\right|^2 = \left(\frac{eA_0}{m_0c}\right)^4 p_0^4\frac{32}{3E_g^2}\xi^4\frac{(4\xi_2^2-1)}{(3+4\xi_2^2)}\left[4e_\perp^2+(4\xi_2^2-1)e'^2_y\right]e'^2_x, \quad (5)$$

we find that the coefficient of two-photon light absorption has the form:

$$K_{c;V}^{(2)} = K_{c;V}^{(2)}(0)e^{\tilde{E}_g/k_BT}e^{-E_V(k_{2\omega})/k_BT}\frac{k_{2\omega}\xi_1}{\frac{k_BT}{E_g}\left(1+\frac{2k_BT}{E_g}\right)}\frac{(4\xi_1^2-1)}{(3+4\xi_1^2)}\left\langle\frac{1}{2\pi}\left[4e_\perp^2+(4\xi_1^2-1)e'^2_y\right]e'^2_x\right\rangle, \quad (6)$$

where $K_{c;\mathrm{V}}^{(2)}(0) = \frac{256}{3}\pi\left(\frac{2\pi e^2}{\hbar n_\omega c}\right)^2\frac{\hbar I\gamma^6 n^{(2)}}{E_g^8}$,$\xi = \hbar\omega/E_g$ and took into account that the law of conservation of energy for a two-photon optical transition.

Fig. 3 shows the spectral and temperature relationships of the interband two-photon absorption coefficient $K_{c;\mathrm{V}}^{(2)}(\omega,T)$ of light in monoatomic layers of transition metal dichalcogenides: a), b), c) pictures show the relationships related to monoatomic layers with forbidden band width $E_g = 0.3\ eV$, $E_g = 0.6\ eV$ and $E_g = 1.0\ eV$ of $K_{c;\mathrm{V}}^{(2)}(\omega,T)$. Lines 1 in the images correspond to linear, and lines 2 correspond to circularly polarized light. In the calculations, the calculated maximum value of $K_{c;\mathrm{V}}^{(2)}(\omega,T=200\ K)$ for $E_g = 0.3\ eV$ was taken as one unit. It can be seen from Figure 3 that both in the spectral connection of the quantity $K_{c;\mathrm{V}}^{(2)}(\omega,T)$ and in the temperature connection, it gradually reaches a maximum with the increase of ω, T, and then decreases. Tables 2 (3) for the exact value of the forbidden zone width of the temperature (frequency) corresponding to the maximum value of $K_{c;\mathrm{V}}^{(2)}(\omega,T)$ at the selected value of the frequency (temperature). Numerical calculations show that: a) the frequency corresponding to the maximum value of $K_{c;\mathrm{V}}^{(2)}(\omega,T)$ at a specific value of the temperature decreases with an increase in the width of the forbidden zone (see Table 2), that is, it reaches a significant value in monoatomic layers with a narrow zone; b) the temperature corresponding to the maximum value of $K_{c;\mathrm{V}}^{(2)}(\omega,T)$ at a specific value of the frequency increases significantly with an increase in the width of the forbidden zone ($T_{\max}$) (see Table 3), that is, in monoatomic layers with a wide zone, it is greater than 1000 K by $T_{\max}$; c) As the bandgap width increases, the maximum value of $K_{c;\mathrm{V}}^{(2)}(\omega,T)$ decreases for both linearly polarized light and circularly polarized light.



a b c

Figure 3. Spectral and temperature correlations of interband two-photon absorption coefficient $K_{c;\mathrm{V}}^{(2)}(\omega,T)$ of light in monoatomic layers of transition metal dichalcogenides: a), b), c) pictures show the correlations $K_{c;\mathrm{V}}^{(2)}(\omega,T)$ of monoatomic layers with forbidden band width $E_g = 0.3\ eV$, $E_g = 0.6\ eV$, and $E_g = 1.0\ eV$. Lines 1 in the figure correspond to linear, and lines 2 correspond to circularly polarized light. In the calculations, the calculated maximum value of $K_{c;\mathrm{V}}^{()}(\omega,T=200\ K)$ for $E_g = 0.3\ eV$ was taken as one unit

Table 2. Calculated results of the frequency corresponding to the maximum values of the two-photon absorption coefficient of light in monoatomic layers of transition metal dichalcogenides versus temperature and bandgap width.

E_g, eV	$\omega_{\max}$ frequencies corresponding to the maximum values of the two-photon absorption coefficient (calculated in units of $E_g/\hbar$)					
	$T = 100\ K$		$T = 300\ K$		$T = 500\ K$	
	Lin. pol.	Cir. pol.	Lin. pol.	Cir. pol.	Lin. pol.	Cir. pol.
1.5	0.514	0.514	0.536	0.557	0.563	0.564

Table 3. Calculated results of the temperature corresponding to the maximum values of the two-photon absorption coefficient of light in monoatomic layers of transition metal dichalcogenides versus frequency and bandgap width.

E_g, eV	$T_{\max}, K$ temperature corresponding to the maximum values of the two-photon absorption coefficient					
	$\hbar\omega = 0.6E_g$		$\hbar\omega = 0.7E_g$		$\hbar\omega = 0.8E_g$	
	Lin. pol.	Cir. pol.	Lin. pol.	Cir. pol.	Lin. pol.	Cir. pol.
1.5	1000	1000	> 1000	> 1000	> 1000	> 1000

CONCLUSIONS

This study presents a theoretical framework for understanding the interband absorption of polarized radiation in monatomic layers of transition metal dichalcogenides (TMDs). Through comprehensive modeling and computational analysis, the spectral and temperature dependencies of one- and two-photon absorption coefficients were examined under linearly and circularly polarized light. The findings reveal that absorption coefficients increase with frequency up to a peak value and then decrease, with the behavior significantly influenced by the material's bandgap width, temperature, and polarization of the incident radiation.

The analysis demonstrated that for a fixed temperature, the frequency corresponding to maximum absorption decreases with increasing bandgap width, suggesting enhanced absorption in narrow-gap TMD layers. Conversely, at a fixed frequency, the temperature corresponding to maximum absorption increases with larger bandgap widths. The results also highlight that the maximum absorption coefficients are more pronounced in narrow-gap TMDs at lower temperatures, emphasizing the role of thermal effects in tailoring optical properties.

These insights contribute to a deeper understanding of the optical transitions in 2D TMD semiconductors and their potential applications in nanoelectronics and optoelectronics. The study provides a foundation for designing advanced photodetectors, light-emitting diodes, and other devices utilizing the unique optical characteristics of monatomic TMD layers. Future work could extend these findings through experimental validation and explore their implications for device optimization under various operational conditions.

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СПЕКТРАЛЬНА ТА ТЕМПЕРАТУРНА ДИНАМІКА ПОГЛИНАННЯ ФОТОНІВ В ОДНОАТОМНИХ ДИХАЛЬКОГЕНІДАХ ПЕРЕХІДНИХ МЕТАЛІВ

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У цьому дослідженні розглядається динаміка міжзонного поглинання поляризованого випромінювання в одноатомних шарах дихалькогенідів перехідних металів (ТМД), що є критично важливим для розвитку нано- та оптоелектроніки. Використовуючи теоретичне моделювання та обчислювальний аналіз, були досліджені спектральні та температурні залежності коефіцієнтів одно- та двофотонного поглинання для лінійно та циркулярно поляризованого світла. Результати показують виражене збільшення коефіцієнтів поглинання з частотою, досягаючи піку перед зниженням, поведінка, що модулюється шириною забороненої зони матеріалу та температурою. Аналіз також підкреслює істотний вплив поляризації світла на характеристики поглинання із залежними від температури зрушеннями максимальної частоти поглинання. Ці знахідки сприяють глибшому розумінню оптичних властивостей матеріалів 2D TMD і відкривають шлях для їх застосування в розробці ефективних фотодетекторів, світлодіодів та інших оптоелектронних пристроїв наступного покоління.

Ключові слова: *одноатомні шари; дихалькогеніди перехідних металів; поглинання фотонів; спектральна залежність; вплив температури; ефекти поляризації*