Structure of the absorption spectrum of a noncubic local center

Yu. E. Perlin, L. S. Kharchenko, and M. Frąckowiak

V. I. Lenin State University, Kishinev

A theory is developed of the profile of an electron-vibrational band representing a transition to a doubly degenerate electronic level of a tetragonal local center. The example of an F (F^-) center in a BaClF crystal is used to show that a calculation of the band profile in the independent ordering approximation yields the results which are in good agreement with the experimental data if use is made of the electron-vibrational interaction constants obtained by an analysis of the moments of the polarization dichroism spectra.

One of the most important manifestations of the Jahn-Teller effect in the spectroscopy of local centers in crystals is a characteristic structure of wide (multiphonon) optical bands that appear as a result of transitions to orbitally degenerate levels. Apart from the factors that depend weakly on the frequency Ω of the absorbed (emitted) light, we shall describe the relevant spectra by the so-called form function[1-3]

$$F(\Omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt\, e^{i\Omega t} I(t) \quad (\hbar = 1), \tag{1}$$

where

$$I(t) = \mathrm{Av}(i) \langle i | e^{iHt} d^+ e^{-iHt} d | i \rangle. \tag{2}$$

Here, H is the Hamiltonian of a system interacting with an electromagnetic radiation field; $|i\rangle$ is the exact wave function of the initial state of this system; d is the electron operator of the transition; $\mathrm{Av}(i)$ represents statistical averaging over thermodynamic-equilibrium populations of the initial states. We shall consider impurity-phonon systems characterized by

$$H = H_e + H_L + H_{eL} = H_0 + H_{eL}, \tag{3}$$

where H_e is the Hamiltonian of the optical electrons of a local center which includes the interaction of these electrons with a defect (with a core in the case of an impurity ion with a partly filled shell) and with a static crystal field;

$$H_L = \sum_\varkappa \omega_\varkappa a_\varkappa^+ a_\varkappa, \tag{4}$$

where the index $\varkappa$ labels the vibrational modes of a crystal (including local modes); $a_\varkappa^+$ and $a_\varkappa$ are the creation and annihilation operators for a phonon of frequency $\omega_\varkappa$; H_{eL} is the Hamiltonian of the electron-phonon interaction which is described by the linear form:

$$H_{eL} = \sum_\varkappa (V_\varkappa a_\varkappa^+ + V_\varkappa^* a_\varkappa), \tag{5}$$

$V_\varkappa$ are the electron operators of the electron-phonon interaction.

If the initial electron-vibrational state is adiabatic, i.e., if this state is not orbitally degenerate and is separated from the other electronic states by an energy gap which is much greater than the maximum phonon energy, the wave function of this state becomes factorized:

$$|i\rangle = |\Gamma_1\rangle \prod_\varkappa |n_\varkappa\rangle. \tag{6}$$

Here, $|\Gamma_1\rangle$ is the electron wave function correponding to a unit irreducible representation of the point group of a local center; $|n_\varkappa\rangle$ are the wave functions of the harmonic oscillators with equilibrium positions corresponding to the adiabatic potential minimum.

Application of the adiabatic form of perturbation theory[1,2,4] makes it possible to include in the electron Hamiltonian the energy of the polaron deformation of the initial state. Then, the renormalized electron-phonon interaction operators have the property

$$\langle \Gamma_1 | V_\varkappa | \Gamma_1 \rangle = 0 \tag{7}$$

and the Hamiltonian H_e describes the Franck-Condon states which correspond to the configuration of the nuclei frozen in the Γ_1 state. Let us assume that Γ is a degenerate electron state of the Hamiltonian H_e. In the case of the $\Gamma_1 \to \Gamma$ transition Eq. (2) becomes

$$I(t) = \exp(-i\Omega_\Gamma t) \sum_{\gamma, \gamma'} \langle \Gamma_1 | d^+ | \Gamma\gamma \rangle$$
$$\times \langle\!\langle \Gamma\gamma | U(t) | \Gamma\gamma' \rangle\!\rangle_L \langle \Gamma\gamma' | d | \Gamma_1 \rangle, \tag{8}$$

where Ω_Γ is the Franck-Condon transition frequency; γ is the row index of the irreducible electron representation Γ; $\langle \ldots \rangle_L$ represents averaging over the phonon occupation numbers;

$$U(t) = T \exp\left\{ -i \int_0^t H_{eL}(t_1)\, dt_1 \right\} \tag{9}$$

is the evolution operator [$T = T_e T_L$ is the chronological ordering symbol and $H_{eL}(t)$ is the electron-phonon interaction operator described in the interaction representation].

The matrices of the operators $V_\varkappa$ and $V_{\varkappa'}$, $(\varkappa' \neq \varkappa)$ derived in the basis of the electron wave functions $|\Gamma\gamma\rangle$ generally do not commute, which is known to lead to the Jahn-Teller effect. However, in the T product they can be treated as commuting quantities. This makes it possible to carry out the exact L averaging[1,2,5] in the case of the linear electron-phonon interaction of Eq. (5):

$$\langle U(t)\rangle_L = T_e \exp[g(t)], \tag{10}$$

where the cumulant function $g(t)$ is of the form

$$g(t) = -\frac{1}{2} \iint_0^t dt_1\, dt_2 \sum_\varkappa V_\varkappa(t_1) V_\varkappa(t_2) \varphi_\varkappa(|t_1-t_2|), \tag{11}$$

$$\varphi_\varkappa(t) = \bar{n}_\varkappa \exp(i\omega_\varkappa t) + (\bar{n}_\varkappa+1)\exp(-i\omega_\varkappa t), \tag{12}$$

$$\bar{n}_\varkappa = (\exp\beta_\varkappa - 1)^{-1}, \qquad \beta_\varkappa = \omega_\varkappa/kT. \tag{13}$$

In the case of small-radius local centers it is convenient to use a group-theoretic representation of the electron-phonon interaction operators of the form[2-4]

$$V_\varkappa = \sum_{\bar{\Gamma}\bar{\gamma}} V_{\bar{\Gamma}\bar{\gamma}} \left(\frac{1}{2\omega_\varkappa N}\right)^{1/2} \langle\varkappa|\bar{\Gamma}\bar{\gamma}\rangle, \tag{14}$$

where $\bar{\Gamma}$ is an irreducible vibrational representation of the point group of the Hamiltonian H_e: $V_{\bar{\Gamma}\bar{\gamma}}$ are the irreducible tensor electron-phonon interaction operators (they are assumed to be real); N is the number of unit cells in a crystal; $\langle\varkappa|\bar{\Gamma}\bar{\gamma}\rangle$ are known as the Van Vleck coefficients forming a matrix for unitary transformation of the symmetrized normal coordinates $Q_{\bar{\Gamma}\bar{\gamma}}$ to the complex normal coordinates $Q_\varkappa$ of the phonon subsystem.

Substitution of Eq. (14) into Eq. (11) gives

$$g(t) = -\frac{1}{2}\int_0^t\int dt_1\, dt_2 \sum_{\bar{\Gamma}\bar{\gamma}} \frac{B_\varkappa(\bar{\Gamma})}{2N\omega_\varkappa} V_{\bar{\Gamma}\bar{\gamma}}(t_1) V_{\bar{\Gamma}\bar{\gamma}}(t_2)$$
$$\times \varphi_\varkappa(|t_1-t_2|). \tag{15}$$

Equation (15) is derived using the Van Vleck unitarity coefficients[2,4] and employing the average

$$B_\varkappa(\bar{\Gamma}) = \frac{1}{[\bar{\Gamma}]} \sum_{\bar{\gamma}} |\langle\varkappa|\bar{\Gamma}\bar{\gamma}\rangle|^2, \tag{16}$$

where $[\bar{\Gamma}]$ represents the dimensions of the $\bar{\Gamma}$ representation.

Further calculations of the form function of a band by means of the cumulant function (15) are not possible without simplifying assumptions. We shall begin by neglecting the dynamic Jahn-Teller effect, i.e., by retaining in the sum over $\bar{\Gamma}$ only those vibrational representations of $\bar{\Gamma}$ for which the $V_{\bar{\Gamma}\bar{\gamma}}$ matrices are diagonal (adiabatic modes). There is no need then for chronological ordering of the electron operators and the transformation gives the form function of the adiabatic approximation, first obtained in Refs. 5 and 6 and subsequently studied by many authors (for a review see Ref. 7). In the case of a strong interaction with nonadiabatic vibrational modes an analytic expression for the form function can be obtained by means of the semiclassical approximation in which the correlation function $\varphi_\varkappa(t)$ is replace with $\varphi_\varkappa(0) = \coth(\beta_\varkappa/2)$. The molecular model of a center derived in Ref. 8 shows that a semiclassical approximation for the $A\to E - e$ and $A\to T - t$ transitions gives rise to two- and three-spike absorption bands, respectively.[1] A characteristic feature of the semiclassical spectra obtained as we go over from an isolated degenerate level is their symmetric profile relative to the frequency Ω_Γ. Such a band profile may be observed at fairly high temperatures, whereas at low temperatures the spectra are characterized by a strong asymme-

try, indicating that the semiclassical approximation is inapplicable at low temperatures. More accurate results are obtained by the independent ordering approximation[9,10] (see also Refs. 2 and 4) based on an approximate separation of the cumulant function:

$$\frac{1}{2}\int_0^t\int dt_1\, dt_2 V_{\bar{\Gamma}\bar{\gamma}}(t_1) V_{\bar{\Gamma}\bar{\gamma}}(t_2) \varphi_\varkappa(|t_1-t_2|)$$
$$\approx \int_0^1\int d\tau_1\, d\tau_2 V_{\bar{\Gamma}\bar{\gamma}}(\tau_1) V_{\bar{\Gamma}\bar{\gamma}}(\tau_2) \int_0^t dt_1 \int_0^{t_1} dt_2 \varphi_\varkappa(t_1-t_2), \tag{17}$$

where τ_1 and τ_2 are the ordering indices.

Substitution of Eq. (17) into Eq. (15) gives

$$g(t) = -\frac{t^2}{4} \sum_{\bar{\Gamma}\bar{\gamma}} \lambda_{\bar{\Gamma}}(t) \left[\int_0^1 d\tau V_{\bar{\Gamma}\bar{\gamma}}(\tau)\right]^2, \tag{18}$$

where

$$\lambda_{\bar{\Gamma}}(t) = \sum_\varkappa \frac{2B_\varkappa(\bar{\Gamma})}{N\omega_\varkappa^3 t^2} [\varphi_\varkappa(0) - \varphi_\varkappa(t) - i\omega_\varkappa t]. \tag{19}$$

Fourier transformation of the expression under the chronological product sign yields

$$\langle U(t)\rangle_L = \int_{-\infty}^\infty \prod_{\bar{\Gamma}\bar{\gamma}} \left[\frac{dq_{\bar{\Gamma}\bar{\gamma}}}{\pi^{1/2}} \exp(-q_{\bar{\Gamma}\bar{\gamma}}^2)\right]$$
$$\times \exp\left[-it \sum_{\bar{\Gamma}\bar{\gamma}} V_{\bar{\Gamma}\bar{\gamma}}\lambda_{\bar{\Gamma}}^{1/2}(t) q_{\bar{\Gamma}\bar{\gamma}}\right]. \tag{20}$$

Equation (20) has the structure of a quantum-statistical average of an exponential operator for a set of effective harmonic oscillators with coordinates $q_{\bar{\Gamma}\bar{\gamma}}$. Since $\langle U(t)\rangle_L$ considered in the basis of the wave functions $|\Gamma\gamma\rangle$ is a multiple of a unit matrix, it follows that

$$\langle\langle\Gamma\gamma|U(t)|\Gamma\gamma'\rangle\rangle_L = \frac{\delta_{\gamma,\gamma'}}{[\Gamma]} \mathrm{Sp}_e\{\langle U(t)\rangle_L\}. \tag{21}$$

In view of the unitary invariance of the trace, its calculation can be carried out in the basis of the eigenfunctions of the operator

$$A(t) = \sum_{\bar{\Gamma}\bar{\gamma}} V_{\bar{\Gamma}\bar{\gamma}}\lambda_{\bar{\Gamma}}^{1/2}(t) q_{\bar{\Gamma}\bar{\gamma}}. \tag{22}$$

In the case of small-radius centers we need in practice consider only a finite number of the coordinates $q_{\bar{\Gamma}\bar{\gamma}}$ governed by the number of the vibrational degrees of freedom of the quasimolecule formed by a local center and by its immediate crystalline environment. Let q_1 be the normal coordinate of a totally symmetric vibration. We shall describe the remaining degrees of freedom introducing a generalized spherical coordinate system which consists of a radial coordinate

$$q = \left[\sum_{\bar{\Gamma}\bar{\gamma}} q_{\bar{\Gamma}\bar{\gamma}}^2\right]^{1/2} \quad (\bar{\Gamma} \neq \Gamma_1)$$

and $p - 1$ angular coordinates $(\vartheta_1, \ldots, \vartheta_{p-1})$.

Using $q\Omega_\alpha(t, \vartheta_1, \ldots, \vartheta_{p-1})$ to denote the eigenvalues of the Hamiltonian (22), we obtain

$$\mathrm{Sp}_e\{\langle U(t)\rangle_L\}=\langle U_1(t)\rangle_{L_1}\sum_\alpha (\pi)^{-p/2}\int d\vartheta \int_0^\infty dq$$

$$\times q^{p-1}\exp[-q^2-iqt\Omega_\alpha(t,\vartheta_1,\ldots,\vartheta_{p-1})], \qquad (23)$$

where $d\vartheta$ represents integration with respect to the angular variables and $U_1(t)$ corresponds to the contribution of the totally symmetric vibrations to Eq. (20). Following Ref. 11, we shall introduce the spectral density of eigenvalues of the operator (22):

$$\rho(\xi)=\int d\vartheta \sum_\alpha \delta(\xi-\Omega_\alpha{}^2). \qquad (24)$$

Then, the generating function of Eq. (2) transforms into

$$I(t)=\frac{1}{[\Gamma]}\sum_\alpha |\langle \Gamma_1|d|\Gamma\gamma\rangle|^2 \exp(-i\Omega_\Gamma t)\langle U_1(t)\rangle_{L_1}$$

$$\times \int d\xi \rho(\xi)(\pi)^{-p/2}\int_0^\infty dq\, q^{p-1}\exp(-q^2-iqt\xi^{1/2}). \qquad (25)$$

Further calculation of the spectrum of a specific system requires the application of a computer. Such a calculation was carried out for the $A_{1g}\to T_{1u}$ $(a+e+t_2)$ transition in the molecular model[11] assuming the same effective phonon frequency for all three vibrational modes and varying the relevant coupling constants. An analogous calculation for the $E\to E(a+e)$ transition was carried out in Ref. 12 on the assumption that $\omega_A=\omega_E$. A comparison with the experimental results was made in Ref. 13, where the calculated profile of the band due to the $^2A_{1g}\to{}^2T_{1u}$ transition was compared with the F absorption band of a CsCl crystal. The function $\lambda_T(t)$ [Eq. (21)] was expanded in Ref. 13 in powers of t up to terms t^3 inclusive. We can easily see that such an expansion including terms up to t^2 brings us back to the semiclassical approximation. Therefore, the results obtained in Ref. 13 represent effectively an asymmetric correction to the semiclassical spectrum, which clearly corresponds to an allowance for the third moment of the absorption band.[14] The agreement between the theory and experiment is obtained by an optimal selection of the electron-phonon interaction constants. Such an agreement is not fully convincing because calculations indicate that very similar spectral profiles are obtained for different sets of these constants. It is more realistic to adopt an approach in which the band profile is calculated in the independent ordering approximation using the electron-phonon interaction constants found independently, for example, from the moments of the polarization dichroism spectra.[2-4] It would be therefore of interest to consider the absorption spectrum of tetragonal $F(F^-)$ centers in crystals of the BaClF type.[15,16] The absorption band due to the $\Gamma_1\to\Gamma_5$ transition (E level in the C_{4v} group) has a strong Jahn-Teller structure. The profile of this band was calculated in Ref. 17 using the semiclassical approximation, but the observed band (Fig. 1) has a strong asymmetry even at room temperature. We shall give below the results of a calculation of the band profile carried out in the independent ordering approximation.
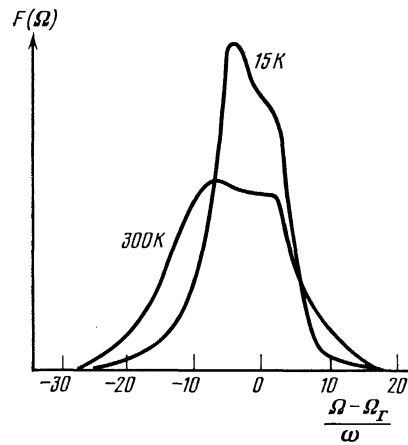

FIG. 1. Absorption band of $F(F^-)$ centers in BaClF crystals (results taken from Refs. 15 and 16).

The problem under consideration is the classical Jahn-Teller problem for the $A\to E(a+b_1+b_2)$ transition. In the quasimolecular model the operator (22) is

$$A(t)=\omega_1 V_1\chi_1{}^{1/2}(t)q_1+\omega_2 V_2\chi_2{}^{1/2}(t)q_2 u_z+\omega_3 V_3\chi_3{}^{1/2}(t)q_3 u_x, \qquad (26)$$

where in the first term we imply the presence of a (2×2) unit matrix; ω_i $(i=1,2,3)$ are the effective frequencies of the $\Gamma_1(a)$, $\Gamma_3(b_1)$, and $\Gamma_4(b_2)$ vibrations, respectively; V_i are the electron-phonon interaction constants; q_i are the effective normal coordinates; u_x and u_z are the Pauli matrices;

$$\chi_i(t)=(2/t^2)[\varphi_i(t)-\varphi_i(0)-i\omega_i t]. \qquad (27)$$

The last relationship means that for each value of Γ in Eq. (19) the frequency ω_x should be replaced with the frequency of the corresponding effective oscillator. In this model the components of the second moment of the F band are

$$\sigma_i=\tfrac{1}{2}V_i{}^2\omega_i{}^2\,\mathrm{cth}(\beta_i/2). \qquad (28)$$

In principle, the change in the third moment of the linear strain and magnetic circular dichroism spectra can be used to determine umambiguously ω_i and the spin-orbit interaction constant λ (Refs. 2–4). However, only the second of these spectra has been obtained for the $F(F^-)$ band,[2] and this has made it possible to find σ_1 and $\sigma_{NS}=\sigma_2+\sigma_3$. The temperature dependences of these quantities were also recorded and these agreed with Eq. (28) on the assumption that $\omega_1=300$ cm^{-1} and $\omega_2=\omega_3=130$ cm^{-1}. In accordance with this result, we shall assume that $\omega_2=\omega_3=\omega$ and $\chi_2=\chi_3=\chi$. Then, the eigenvalues of the operator (26) are described by the following expression if we redefine the variable ξ:

$$\Omega_{1,2}=\pm\chi(t)\omega\xi^{1/2},$$
$$\xi=[V_2{}^2\cos^2\vartheta+V_3{}^2\sin^2\vartheta]^{1/2}, \quad \vartheta=\mathrm{arctg}(q_3/q_2). \qquad (29)$$

Substitution of Eq. (29) into Eq. (24) gives

$$\rho(\xi)=\theta(V_2{}^2-\xi)\theta(\xi-V_3{}^2)/\pi[(V_2{}^2-\xi)(\xi-V_3{}^2)]^{1/2}, \qquad (30)$$

where θ is the step function.

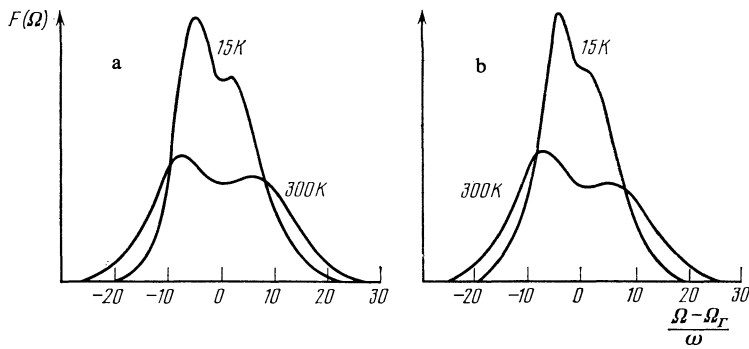After substitution of $\chi(t)$ from Eq. (27) into Eq. (25), we obtain

FIG. 2. Form function of an absorption band for the due to the $A \to E(a + b_1 + b_2)$ transition: a) $V_1^2/V_2^2 = 1.5$; b) $V_1^2/V_2^2 = 4$.

$$F_{NS}(\Omega) = \int d\xi \rho(\xi) \left\{ \delta(\Omega - \Omega_r) + \xi \left[-\operatorname{cth} \frac{\beta}{2} f(\Omega, \xi) \right. \right.$$

$$\left. \left. + \omega \frac{\partial}{\partial \Omega} f(\Omega, \xi) + (\bar{n} + 1) f(\Omega - \omega, \xi) + \bar{n} f(\Omega + \omega, \xi) \right] \right\}, \quad (31)$$

$$f(\Omega, \xi) = \sum_{n=-\infty}^{\infty} e^{n\beta/2} \int_0^1 d\tau \exp\left\{ -\frac{\xi}{2} (1 - \tau^2) \operatorname{cth} \frac{\beta}{2} \right\}$$

$$\times I_n \left[\frac{\xi(1 - \tau^2)}{2 \operatorname{sh}(\beta/2)} \right] \delta\left(\Omega - \Omega_r + \frac{\xi}{2} (1 - \tau^2) \omega - n\omega \right), \quad (32)$$

where $I_n(z)$ is a modified Bessel function.

If in Eq. (20) we retain only the factor representing the totally symmetric vibrations, then in the adopted approximation of the effective frequencies we obtain the form function $F_S(\Omega)$ which is exactly identical with the results of Refs. 5 and 6. A complete spectral distribution is now easily calculated from the convolution formula

$$F(\Omega) = \int_{-\infty}^{\infty} F_S(\Omega - \Omega') F_{NS}(\Omega') d\Omega'. \quad (33)$$

The independent ordering approximation gives (like the semiclassical approximation) only the envelope of the spectrum and not its detailed behavior within the limits of one vibrational frequency. Consequently, the calculation program includes averaging of the form function within these limits at each frequency of light.

Figure 2 shows the graphs of the form function $F(\Omega)$ at temperatures of 15 and 300 K for the values of ω_1 and ω given above; $V_1^2 = 4.4$ and $V_2^2 + V_3^2 = 50$ (Ref. 15), where V_2^2 and V_3^2 were varied within the limits of this sum. The resultant two-peak spectra resemble qualitatively the spectra of the $A \to E \cdot (a + e)$ band taken from Ref. 12.

However, the spectral profile depends strongly on the absolute values of V_3 and V_2 and on their ratio: as V_2/V_3 increases, the peaks come closer together and for $V_3 = 0$ the spectrum should become bell-shaped. On increase in the temperature of a crystal the separation between the peaks increases, but even at $T = 300$ K the spectrum remain asymmetric.

The calculated spectrum which agrees best with the experimental results is shown in Fig. 2b. Naturally, complete agreement between the calculated and observed band pro-

files cannot be expected because of the approximations described above and also because mixing of the excited states of the F center by the lattice vibrations makes some contribution to the absorption band parameters. The calculated separations between the peaks in the band are 850 cm^{-1} ($T = 15$ K) and 2000 cm^{-1} ($T = 300$ K), whereas the experimental results[15,16] give 800 and 2000 cm^{-1}, respectively.

It follows that the independent ordering approximation in combination with determination of the electron-phonon interaction constants from the moments of the polarization dichroism spectra makes it possible to calculate quantitatively the spectra of various Jahn-Teller centers without fitting parameters. In the above calculations the values of V_2 and V_3 had to be selected because of the absence of comprehensive experimental data on the polarization dichroism. In this situation the proposed method provides an additional opportunity for determining the electron-phonon interaction constants.

Sometimes one encounters an experimental situation in which the observed structure of an absorption band can only be tentatively attributed to the Jahn-Teller effect; an alternative cause may be splitting of the degenerate electron levels because of a purely electronic perturbation. An example of this kind is an excited state of the V^{2+} ion in MgO (Ref. 18). We are of the opinion that the application of the above method would be decisive in such a situation. In particular, in the case of the $F(F^-)$ center in BaClF the "Jahn-Teller" nature of the band (Fig. 1) can be regarded as proven.

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[1]We shall use not only the notation Γ_i for the irreducible representations, but also the notation A, E, and T for one-, two-, and three-dimensional electronic representatives and we shall identify the corresponding vibrational representations by lower-case letters.

[2]According to the estimates in Ref. 15, the contribution of the spin-orbit interaction to the second moment of an absorption band is less than 1% and it is ignored in our calculations.

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