

Modulation broadening of spectral lines for impurity centers with degenerate levels

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Modulation broadening is considered for spectral lines corresponding to degenerate high-frequency localized vibrations and electronic transitions. It is found that the processes occurring in fourth-order perturbation theory with regard to the interaction linear in the phonon operators, which mixes impurity states having the same energy, lead to a T^5 modulation broadening at temperatures much below the Debye temperature (instead of T^7 for nondegenerate levels). Detailed consideration is given to the case of threefold degenerate dipole-active localized vibrations. The line widths at the fundamental frequency and the harmonic are related in agreement with experiment. The influence of this mechanism of modulation broadening on the self-induced change of polarization is studied, for radiation at a frequency close to the natural frequency of the impurity transition.

The narrow lines in the optical spectra of impurity centers in crystals may be due either to transitions between different electron states¹ or to localized vibrations.² If the frequencies of the relevant transitions are much greater than the phonon frequencies, then at low temperatures the zero-phonon lines are extremely narrow (if there is no inhomogeneous broadening), but their widths increase rapidly with rising temperature. The basic mechanism of high-frequency line broadening is usually a modulation mechanism,³⁻¹⁰ involving phase fluctuations of the impurity center wave function, due to phonon scattering by the center. The modulation broadening is usually associated with terms in the impurity excitation-phonon interaction Hamiltonian that are quadratic in the phonon operators.

Since, as the temperature rises, the occupation numbers of the phonons scattered increase rapidly, so does the modulation broadening,^{5,6,8,10} as $(T/\omega_D)^2$ for $T \geq \omega_D$ and as $(T/\omega_D)^7$ for $T \ll \omega_D$ (where ω_D is the Debye frequency; $\hbar = k = 1$). For impurities with nondegenerate levels, the interaction linear in the phonon operators does not affect the temperature dependence of the modulation broadening. It simply alters the quadratic interaction in a way proportional to the ratio of the squared linear interaction constant to the distance between impurity levels, corresponding to the nonadiabatic approximation.

For impurities with degenerate levels, the role of the linear interaction is much more significant even for weak coupling (when the binding energy is much less than the characteristic phonon energies). Since it mixes the impurity quantum states that have equal energies, the corresponding change in the quadratic interaction parameters is proportional to the ratio of the squared coupling constant to the phonon frequency (which is considerably less than the distance between levels). It will be shown below that for $T \ll \omega_D$ the modulation broadening then varies not as $(T/\omega_D)^7$ but as $(T/\omega_D)^5$, i.e., is considerably greater than for nondegenerate levels. Note also that the number of modulation broadening constants is greater than in the case usually considered, where the Hamiltonian of the impurity excitation-phonon interaction quadratic in the phonon operators depends only on the atomic

displacements (and is the same as when the dependence of the interaction both on the displacements and on the velocities is taken into account). This is reflected not only in the line widths but also in the relationship between the line widths at localized vibration harmonics, and also in the nonlinear resonant response of the impurity and in the nature of the self-induced change in the polarization of the resonance radiation propagating in the crystal.

1. MODULATION BROADENING DUE TO THE INTERACTION LINEAR IN THE PHONON OPERATORS

Let us first consider the change in the modulation broadening for a degenerate localized vibration. The Hamiltonian of the localized vibration-crystal system, neglecting the internal anharmonicity of the vibration (see below), is

$$H = H_0 + H_m + H_i, \quad H_0 = \omega, \quad \sum_k a_k^\dagger a_k, \quad H_m = \sum_k \omega_k b_k^\dagger b_k, \quad (1)$$

$$H_i = H_i^{(3)} + H_i^{(4)}, \quad H_i^{(3)} = \sum_{xx'k} (V_{xx'k} a_k^\dagger a_x b_k + \text{H.c.}),$$

$$H_i^{(4)} = \sum_{xx'kk'} V_{xx'kk'} a_k^\dagger a_x b_k^\dagger b_{k'}. \quad (2)$$

Here, the subscript κ labels the degrees of freedom of the degenerate localized vibration; k labels the continuous-spectrum vibrations; a_κ , b_k , are Bose operators for the respective vibrations. Since high-frequency vibrations ($\omega_0 \gg \omega_k$) are under consideration, only the "adiabatic" terms are kept in the anharmonic interaction Hamiltonian, which relate localized vibration states with the same energy (apart from the multiplet level splittings). When the interaction depends only on the displacements of the atoms, i.e., on the operators $a_\kappa + a_\kappa^\dagger$, $b_k + b_k^\dagger$, we evidently have

$$V_{xx'k} = V_{x'xk} = V_{xx'k}^*, \quad V_{xx'kk'} = V_{x'xkk'} = V_{xx'k'k} = V_{xx'kk'}^*, \quad (3)$$

The localized vibration optical spectrum profile near the main peak, where $\omega \approx \omega_0$ and the phonon wings can be neglected, is represented by $\text{Im} \ll a_\kappa, a_\kappa^\dagger \gg_{\omega+i0}$, where $\ll a_\kappa, a_\kappa^\dagger \gg_{\omega+i0}$ is the Fourier transform of the retarded Green function. The equation of motion for this function is

$$\ll a_\kappa, a_\kappa^\dagger \gg_{\omega - \omega_0} = 1 + \sum_{x'k} (V_{xx'k} \ll a_\kappa, b_k, a_\kappa^\dagger \gg + V_{xx'k}^* \ll a_\kappa, b_k^\dagger, a_\kappa^\dagger \gg)$$

$$+ \sum_{k \neq k'} V_{xx'kk'} \langle a_{x'} b_{k'}^{\dagger} a_x \rangle.$$

The terms with $k = k'$ in this expression modify the frequency ω_0 , which we will call the produced frequency; those with $k \neq k'$ in second-order perturbation theory determine the modulation broadening as usually considered. The terms in Eq. (4) due to the Hamiltonian $H_1^{(3)}$ in system (2) and proportional to $V_{\kappa\kappa'k}$ also cause modulation broadening, but in higher orders of perturbation theory. Retaining in the equations for these Green functions only the terms having the same form as the last one in Eq. (4) (and omitting the terms causing line shift and decay processes), we find that the second order the interaction $H_1^{(3)}$ changes the last term in Eq. (4) as follows:

$$\begin{aligned} \tilde{V}_{xx'kk'} &= V_{xx'kk'} + \sum_{x_1} \left[\frac{V_{x_1xk} V_{x_1x'k'}}{\omega - \omega_{x_1} + \omega_k} + \frac{V_{xx'k'} V_{x_1x_1k}}{\omega - \omega_{x_1} - \omega_k} \right] = V_{xx'kk'} \\ &+ \sum_{x_1} \left[\frac{1}{\omega_k} V_{x_1xk} V_{x_1x'k'} - \frac{1}{\omega_{k'}} V_{xx'k'} V_{x_1x_1k} \right]. \end{aligned} \quad (5)$$

The second equation here uses the fact that the modulation broadening is considerably less than the phonon frequency, and in the denominator small terms $\omega - \omega_{\kappa 1}$ have been neglected in comparison with ω_k and $\omega_{k'}$.

The solution of Eq. (4) in the second order with respect to the parameters $\tilde{V}_{\kappa\kappa'}$ then gives the following expressions for the Green function $\langle a_{\kappa}, a_{\kappa'}^{\dagger} \rangle$ and the modulation broadening $\Gamma^{(1)}$ at the fundamental frequency:

$$\langle a_{\kappa}, a_{\kappa'}^{\dagger} \rangle_{\omega+i0} = [\omega - \omega_0 - i\Gamma^{(1)}]^{-1}, \quad (6a)$$

$$\Gamma^{(1)} = \Gamma_{m1} + 2\Gamma_{m2}, \quad \Gamma_{m1} = \sum_{kk'} |\tilde{V}_{xx'kk'}|^2 \mathcal{S}_{kk'},$$

$$\Gamma_{m2} = \frac{1}{2} \sum_{\substack{x' (x' \neq x) \\ kk'}} |\tilde{V}_{xx'kk'}|^2 \mathcal{S}_{kk'}, \quad \mathcal{S}_{kk'} = \pi (1 + \bar{n}_k) \bar{n}_{k'} (\omega_k - \omega_{k'}), \quad (6b)$$

$$\bar{n}_k = [\exp(\omega_k/T) - 1]^{-1}. \quad (6c)$$

The parameter Γ_{m1} represents phonon scattering which shifts the κ vibration wave function phase; Γ_{m2} represents phonon scattering which converts the κ vibration into another one κ' with the same energy but $\kappa' (\kappa' \neq \kappa)$.

For interaction with long-wave acoustic phonons, which make the main contribution to the modulation broadening (6) at low temperatures, the interaction parameters $V_{\kappa\kappa'kk'} \propto (kk')^{1/2}$. This gives $\Gamma \sim T^7$ when $T \ll \omega_D$. For interaction with these phonons, $V_{\kappa\kappa'k} \sim k^{1/2}$, however. Accordingly, the second term in $V_{\kappa\kappa'kk'}$, Eq. (5), for $\omega_k = \omega_{k'}$ is independent of k [the terms with $\omega_k = \omega_{k'}$ are those which contribute to Γ_{m1} and Γ_{m2} , system (6)]. In the nondegenerate case ($\kappa = \kappa_1 = \kappa'$) this term is zero. In the degenerate case, it is much greater than $V_{\kappa\kappa'kk'}$ in modulus. It readily follows that the temperature dependence of Γ_m for a degenerate localized vibration when $T \ll \omega_D$ has a T^5 form and can easily be observed (when $T \gtrsim \omega_D$, $\Gamma_m \sim T^2$, as in the nondegenerate case). There is a similar temperature dependence¹¹ for the Raman broadening in low-frequency two-level systems with a transition frequency much less than T and ω_D .

The Hamiltonian (1) also describes the kinetics of a two-level electron transition with a degenerate excited level and interacting with phonons; ω_0 here

(4) denotes the transition frequency, $a_{\kappa}^{\dagger}, a_{\kappa}$ the projection operators for the excited state κ . In general, one has also to include, in this case, the Hamiltonian terms that correspond to the electron-phonon interaction in the ground state, but their effect amounts to changing the interaction constants $V_{\kappa\kappa'k'}$, $V_{\kappa\kappa'kk'}$ and ω_0 . When the excited state occupation is negligible, the expression for the modulation broadening of the electron transition optical spectrum is the same as in system (6). For two-dimensional electron systems with a translationally degenerate discrete energy spectrum in a magnetic field (Landau levels), the relaxation due to processes of the type considered, in the fourth order with respect to $H_1^{(3)}$, has been discussed.¹²

2. MODULATION BROADENING FOR THREEFOLD DEGENERATE DIPOLE-ACTIVE LOCALIZED VIBRATIONS WITH CUBIC SYMMETRY

One of the most thoroughly studied types of localized vibrations is threefold degenerate ones with cubic symmetry.¹³ In the harmonic approximation, these are a set of vibrations with the same frequencies and dipole moments oriented along $\langle 100 \rangle$ type crystal axes. The Hamiltonian of isolated localized vibrations in this approximation is represented by H_0 in Eq. (1), κ taking the values x, y, z . The localized vibration energy levels, if anharmonicity is neglected, are $(n+1) \cdot (n+2)/2$ -fold degenerate, where n is the principal quantum number. When the anharmonicity is taken into account, these levels split into multiplets.¹⁴ The distances between levels in the low multiplets are usually much less than that between the multiplets ($\sim \omega_0$) and the crystal Debye frequency ω_D .

To represent the peak profiles in the optical spectra of localized vibrations, it is convenient to use a treatment developed previously^{15,16} to derive the quantum kinetic equation in operator form for the density matrix ρ of an anharmonic localized vibration. It can be shown that for degenerate localized vibrations the term in the equation for ρ (in the collision integral) which describes the modulation broadening is

$$\partial \rho / \partial t = -\hat{\Gamma}_m \rho, \quad (7a)$$

$$\hat{\Gamma}_m = \sum_{x_1 x_2 x_3 x_4} \Gamma_{x_1 x_2 x_3 x_4} [a_{x_1}^{\dagger} a_{x_2}, [a_{x_3}^{\dagger} a_{x_4}, \rho]]. \quad (7b)$$

The relaxation parameters $\Gamma_{\kappa_1 \kappa_2 \kappa_3 \kappa_4}$ are seen from Eq. (7b) to be the components of a rank-four tensor symmetric under interchange of pairs of subscripts (but not the subscripts in a pair). In the case considered, a threefold degenerate localized vibration with cubic symmetry, this tensor has four independent components; the operator describing the modulation broadening is

$$\begin{aligned} \Gamma_m \rho &= \Gamma_{m1} \sum_x [(a_x^{\dagger} a_x)^2 \rho - 2a_x^{\dagger} a_x \rho a_x^{\dagger} a_x + \rho (a_x^{\dagger} a_x)^2] + \Gamma_{m2} \sum_{xx'} [a_x^{\dagger} a_x a_{x'} a_{x'}^{\dagger} \rho \\ &- 2a_x^{\dagger} a_x \rho a_{x'}^{\dagger} a_{x'} + \rho a_x^{\dagger} a_x a_{x'} a_{x'}^{\dagger}] + \Gamma_{m3} \sum_{xx'} [a_x^{\dagger} a_x a_{x'}^{\dagger} a_{x'} \rho - 2a_x^{\dagger} a_x \rho a_{x'}^{\dagger} a_{x'} \\ &+ \rho a_x^{\dagger} a_x a_{x'}^{\dagger} a_{x'}] + \Gamma_{m4} \sum_{xx'} [(a_x^{\dagger} a_x)^2 \rho - 2a_x^{\dagger} a_x \rho a_x^{\dagger} a_x + \rho (a_x^{\dagger} a_x)^2], \end{aligned} \quad (8)$$

where

$$\Gamma_{m3} = \sum_{kk'} \tilde{V}_{xx'kk'} \tilde{V}_{x'x'kk'} \mathcal{S}_{kk'}, \quad \Gamma_{m4} = \sum_{kk'} \tilde{V}_{xx'kk'} \tilde{V}_{xx'kk'} \mathcal{S}_{kk'}, \quad (x \neq x') \quad (9)$$

the prime to the sums in Eq. (8) signifies that the terms with $\kappa = \kappa'$ are to be omitted.

In the particular case $\Gamma_{m4} = \Gamma_{m2}$, the expression (8) agrees with that found previously¹⁶; it is seen from Eqs. (5) and (8) that this condition is satisfied when $\hat{V}_{\kappa\kappa'kk'} = \hat{V}_{\kappa'\kappa kk'}$, which occurs if no account is taken of the change (5) in the parameters $V_{\kappa\kappa'kk'}$ and the interaction of the localized vibrations with the lattice depends only on the displacements of atoms.

The significance of the parameters Γ_{m1} and Γ_{m2} appearing in Eq. (8) and defined in system (6) has been explained above. The terms in Γ_{m3} and Γ_{m4} in Eq. (8) arise from the interference of phonon scattering by different localized vibration states with equal energies. Accordingly, the parameters Γ_{m3} and Γ_{m4} do not have a definite sign (though they are real by symmetry). It follows from Eqs. (6) and (9) with the Cauchy inequality that

$$\Gamma_{m3} \leq \Gamma_{m1}, \quad \Gamma_{m4} \leq \Gamma_{m2}. \quad (10)$$

When the localized vibration-phonon interaction depends only on the coordinates, so that the conditions (3) are satisfied, at low temperatures $\Gamma_{m2} = -\Gamma_{m4} \propto T^5$, according to Eq. (5); Γ_{m1} , $\Gamma_{m3} \propto T^7$ and are relatively small ($\Gamma_{m1}, \Gamma_{m3} \ll \Gamma_{m2}, |\Gamma_{m4}|$).

It is easy to see that, when the occupation of the localized vibration excited state is negligible Eq. (8) gives system (6); the Green function $\langle\langle a_{\kappa}, a_{\kappa}^+ \rangle\rangle_{\omega+i0}$ is determined by the Fourier transform,

$$(a_{\kappa} \rho(t)) \sim \exp[-(\Gamma_{m1} + 2\Gamma_{m2})t].$$

Other characteristics of the optical spectra accessible to direct measurements (along with the fundamental frequency line width) are governed by combinations of the Γ_{mi} ($i = 1 \dots 4$).

Let us first consider the relationship of the absorption line widths at the fundamental and harmonics of high-frequency localized vibrations, which have been studied experimentally¹⁴ for impurity centers with symmetry T_d (H^- ions in calcium fluoride). The half-width of the dipole absorption line at twice the frequency of the corresponding localized vibrations is known^{13,14} to be determined by the time decay of the dipole moment quadratic in the coordinates of the localized vibrations, that is, by the time decay of $\text{Tr}(a_{\kappa} a_{\kappa'} \rho(t))$, ($\kappa \neq \kappa'$) or, equivalently, the imaginary part of the polarization operator of the Green function $\langle\langle a_{\kappa} a_{\kappa'}^+, a_{\kappa'}^+ a_{\kappa} \rangle\rangle_{\omega+i0}$.

It follows from Eq. (8) that, if the occupation of the localized vibration excited levels is neglected, the line half-width corresponding to the transition to the T state is

$$\Gamma^{(2T)} = 2\Gamma_{m1} + 6\Gamma_{m2} + 2\Gamma_{m3}. \quad (11)$$

This agrees with the previous result.¹⁷

From Eq. (8), we get for the lines corresponding to dipole-forbidden transitions at twice the frequency¹⁾ to the nondegenerate completely symmetric state A and to the doubly degenerate E term, respectively,

$$\Gamma^{(2A)} = 4(\Gamma_{m1} + \Gamma_{m2} + \Gamma_{m4}), \quad \Gamma^{(2E)} = 4(\Gamma_{m1} + \Gamma_{m2}) - 2\Gamma_{m4}. \quad (12)$$

We can also give expressions for the half-widths of lines corresponding to dipole-allowed transitions at three times the localized vibration frequency. These depend on the decay of $\text{Tr}(a_{\kappa}^3 \rho)$ and $\text{Tr}[a_{\kappa'} (a_{\kappa'}^2 + a_{\kappa'}^2) \rho]$, ($\kappa' \neq \kappa'' \neq \kappa$).

As a result of the localized vibration level splitting due to internal anharmonicity, the spectrum contains two dipole transitions at a frequency $\approx 3\omega_0$. The wave functions of the corresponding excited levels are

$$\psi_{xi} = \frac{1}{\sqrt{6}} \cos \theta_i (a_{\kappa'}^2 |0\rangle + 1/2 \sin \theta_i [a_{\kappa'}^2 (a_{\kappa'}^2)^2 + (a_{\kappa''}^2)^2] |0\rangle), \quad (13a)$$

$$i = 1, 2; \quad \kappa' \neq \kappa'' \neq \kappa;$$

$$\kappa = x, y, z; \quad \theta_2 = \theta_1 + \frac{\pi}{2}, \quad (13b)$$

where $|0\rangle$ is the ground state. The quantities θ_i are found by solving the secular equation obtained by diagonalizing the Hamiltonian of an isolated degenerate localized vibration with internal anharmonicity, and have been determined¹⁴ for specific values of the parameters. For transitions to states with $i = 1, 2$, the half-widths are

$$\Gamma_m^{(3,i)} = 3 \cos^2 \theta_i (3\Gamma_{m1} + 2\Gamma_{m2}) + 2\sqrt{6} \sin 2\theta_i \Gamma_{m4} + \sin^2 \theta_i (5\Gamma_{m1} + 10\Gamma_{m2} + 4\Gamma_{m3} + 2\Gamma_{m4}). \quad (14)$$

The measured¹⁴ relationship of the localized vibration absorption line widths in the $\text{CaF}_2:H^-$ system at frequencies $\approx 2\omega_0$ and $\approx \omega_0$ at 100 K is ≈ 2.5 . If we assume that the main contribution to the widths $\Gamma^{(1)}$ and $\Gamma^{(2T)}$ of dipole-active transitions at the fundamental and double frequencies comes from the modulation mechanism, and the interaction depends only on the atomic displacements, and the condition (3) is satisfied, so that $\Gamma_{m1}, |\Gamma_{m3}| \ll \Gamma_{m2} \propto T^5$, then it follows immediately from Eqs. (6) and (11) that $\Gamma^{(2T)}/\Gamma^{(1)} = 3$. The agreement with the experimental results can be regarded as entirely satisfactory, especially if one remembers that in the experiment the absorption line width at the fundamental frequency is fairly strongly exaggerated by the resolution of the apparatus. This also prevents a correlation with experiment at lower temperatures. The correlation is still further hampered for the line at frequency $\approx 3\omega_0$, since in that case the distances between levels in the multiplet¹⁴ are comparable with the characteristic phonon frequencies, and therefore decay processes are important in contributing to the line widths.

To separate the contributions of different mechanisms to the line broadening and determine the modulation broadening parameters for localized vibrations and electronic centers with degenerate levels, it is convenient to use nonlinear optical resonance effects, namely absorption saturation¹⁸ and self-induced change in light polarization in crystals.¹⁹ The absorption saturation is due to equalization of the populations of the levels between which optical transitions take place; the self-induced polarization change is due to the effect on polarized radiation propagation of the anisotropy induced by the radiation (in particular, the different occupation of impurity states degenerate as regards energy). Absorption saturation has been observed²⁰ for localized vibrations in the $\text{CaF}_2:H^-$ system.

A theory of nonlinear resonant response and self-induced polarization change for a two-level impurity where the ground state is completely symmetric and the excited state is transformed by the vector representation of the cubic group has been evolved¹⁹; it applies also to high-frequency localized vibrations with fairly strong anharmonicity, where the transitions from the $n = 1$ level to the multiplet levels with $n = 2$ are nonreso-

nant. No account was taken ¹⁹ of the difference between Γ_{m4} and Γ_{m2} due to the change in the interaction with phonons, Eq. (5). It can be shown with Eq. (8) that the qualitative results and the general expressions ¹⁹ for the nonlinear susceptibilities which determine the absorption saturation and the self-induced polarization change remain valid. But the parameters a and Γ_ϕ in these expressions become

$$a = (\Gamma_{m1} - \Gamma_{m2} - \Gamma_{m3} - \Gamma_{m4})/(\Gamma + 3\Gamma_{m2}), \quad (15)$$

$$\Gamma_\phi = (\Gamma + \Gamma_{m1} + 2\Gamma_{m2} - \Gamma_{m3} - \Gamma_{m4}).$$

where 2Γ is the reciprocal lifetime of the excited state.

The parameter a determines the anisotropy of the nonlinear polarizability of the system. When $a > 0$, as the linearly polarized resonance radiation propagates in the [001] direction in the impure crystal, the electric field vector E (the polarization plane) rotates to the [100] or [010] direction nearest to its original direction; when $a < 0$, it rotates to the nearest $\langle 110 \rangle$ direction.

When the conditions (3) are satisfied, for $T < \omega_D$, as already noted, $\Gamma_{m4} \approx -\Gamma_{m2}$ and $\Gamma_{m2} \gg \Gamma_{m1}$, $|\Gamma_{m2}|$, and from Eq. (15) $a \ll 1$. The polarization plane rotation is then small and increases rapidly (as T^2) with the temperature; when $T \sim \omega_D$, a becomes ~ 1 and reaches saturation.

The sign and magnitude of Γ_{m4} can be determined directly in the realistic case $\Gamma_{m2} \gg \Gamma$ using the change in the polarization of elliptically polarized radiation propagating in the [001] direction. It can be shown that when $\Gamma_{m4} \neq 0$ the polarization change occurs even when the radiation frequency is in the center of the impurity absorption line. When $\Gamma_{m4} > 0$, the phase difference of the components E_y and E_x [with x along (100) and y along (010)] approaches the nearest value πn , $n = 0, \pm 1, \dots$ (corresponding to linear polarization); when $\Gamma_{m4} < 0$, it approaches a $(1/2)\pi(2n + 1)$; that is, the axis of the polarization ellipse approaches a $\langle 100 \rangle$ type axis.

The self-induced polarization change for resonance radiation is thus very sensitive to the modulation broadening parameters and in principle allows all of these to be determined. In this respect, it would be interesting to make an experimental study of the self-induced polarization change for degenerate weakly coupled centers.

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¹⁾ It is assumed that the multiplet level splitting due to the localized vibration anharmonicity is much greater than the line widths, so that the lines corresponding to different transitions are well resolved. The expressions (7) and (8) are then valid so long as the distances between the multiplet levels are less than $\min(T, \omega_D)$.

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