

DYNAMICS OF THE PARAELASTIC TUNNELING SYSTEM
(XY₈) IN THE INTERMEDIATE COUPLING REGIME

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Paraelastic tunneling system (XY₈), in which the paraelastic center (PEC) can occupy one of the 8 off-center sites of the type [111] around some lattice point of the cubic crystal, is considered. The energy eigenvalues of the whole system and the transition rates of the center between its energy eigen-states are calculated.

An important property of paraelastic systems is strong coupling of the center with the lattice vibrations of the crystal. This causes great deformations of the lattice in the neighbourhood of the center and leads to the renormalisation of the tunneling matrix elements. The center must be treated as a small polaron. A satisfactory theory exists for the case when the small-polaron binding energy of the center is much greater than the tunneling matrix elements, that is, when the localisation of the center in one of the off-center positions is very strong. We are interested, on the contrary, in the intermediate case when the tunneling matrix elements are comparable in size with the small-polaron binding energy.

To simplify the description of our system, we take the Abelian group D_{2h} as the symmetry group of the hamiltonian, rather than the non-Abelian O_h which describes its complete symmetry. It is then possible to eliminate in the Schrödinger equation the coordinates of the center, thus obtaining¹⁾

$$\hat{H}_{ph} |f_m(n)\rangle = E(m) |f_m(n)\rangle \quad (1)$$

$$\hat{H}_{ph} = - \sum_{\hat{G}1,1} w(\hat{G}1,1) \chi_m(G) \prod_{s\lambda} [\chi_s(G)]^{n_{s\lambda}} a_{s\lambda}^+ a_{s\lambda} + \sum_{s\lambda} \omega_{\lambda} a_{s\lambda}^+ a_{s\lambda} - \sum_{s\lambda} \omega_{\lambda} (z_{s\lambda} a_{s\lambda}^+ + z_{s\lambda}^* a_{s\lambda}).$$

\hat{H}_{ph} is the new hamiltonian that depends only on the phonon coordinates, $|f_m(n)\rangle$ is the projection of the wave function of the whole system on the localized state $|1\rangle$ of the PEC in the $[111]$ direction, m denoting its symmetry and n the state of the lattice, and $E(m)$ is the energy of the system. Further, we have denoted by G the symmetry operations of the group D_{2h} , $m, s = 1-8$ specify the 1-dimensional irreducible representations, and $\chi_m(G) = \pm 1$ are the group characters. ω_{λ} is the frequency of the mode λ , while $a_{s\lambda}^+$ and $a_{s\lambda}$ denote, respectively, the creation and annihilation operators of symmetry s for this mode. $w(\hat{G}1,1)$ is the tunneling matrix element connecting the localized states $|1\rangle$ and $|\hat{G}1\rangle$ of the center, $z_{s\lambda}$ are the symmetrized coupling constants, and we have set $\hbar = 1$.

We try to solve eq.(1) by a linear combination of coherent states

$$|f_m(n)\rangle = \sum_G c(G) \chi_m(G) \prod_{s\lambda} [\chi_s(G)]^{n_{s\lambda}} |f^G(n)\rangle, \quad (2)$$

where $c(G)$ are expansion coefficients, $n_{s\lambda}$ the number of phonons in the mode s, λ , and $|f^G(n)\rangle$ the coherent states defined as $|f^G(n)\rangle = S(\{x_{s\lambda} \chi_s(G) z_{s\lambda}\}) |n\rangle$, $S(\{z_{s\lambda}\}) = \exp \left\{ \sum_{s\lambda} (z_{s\lambda} a_{s\lambda}^+ - z_{s\lambda}^* a_{s\lambda}) \right\}$. $x_{s\lambda}$ are variational parameters describing the magnitude of the lattice deformations around the PEC and are to be determined by minimizing the energy of the system. By solving eq.(1), while neglecting the terms containing the overlap integrals $\langle f^G(n) | f^{G'}(n) \rangle$, $G' \neq G$, we obtain for the energy of the system

$$E(m) = \sum_{s\lambda} \omega_{\lambda} n_{s\lambda} + [x^2 - 2x - 2(1-x)(2x - \epsilon a)(3x - 1)^{-1}] E_b. \quad (3)$$

Here, E_b stands for the small-polaron binding energy, $a = 3w/2E_b$, where only the tunneling matrix element w for the nearest-neighbour sites is taken into account, and $\epsilon = \pm 1$

for even and odd symmetry m , respectively. The variational parameters $x_{s\lambda} \equiv x$ turn out to be independent of s, λ .

The interaction of PEC with phonons couples only the states of the center of the same symmetry. On acting by \hat{H}_{ph} on $|f_m(n)\rangle$, while retaining the terms that contain the overlap integrals, we obtain for the residual interaction

$$\hat{H}_{res} |f_m(n)\rangle = \sum_G \sum_{s\lambda} \omega_c(G) \chi_m(G) \prod_{s\lambda} [\chi_s(G)]^{n_{s\lambda}} [x \chi_s(G) - 1] \cdot S(\{x \chi_s(G) z_{s\lambda}\}) (z_{s\lambda} a_{s\lambda}^+ + z_{s\lambda}^* a_{s\lambda}) |n\rangle. \quad (4)$$

We calculate the transition rates in the first order perturbation theory. As we are not interested in the states of the lattice, we sum over all possible final states $|n'\rangle$ and take the thermodynamic average over the initial states $|n\rangle$. In the approximation $\alpha \ll 1$ the probabilities for all the allowed transitions of the center from a lower to a higher energy state are equal. We write down the transition rate for such a process in the Debye approximation and in the limit $t \rightarrow \infty$:

$$P = \frac{1}{4\pi} x^2 (3x-1) \left(\frac{\Delta U}{kT_0}\right)^2 \frac{\Delta U}{e^{\Delta U/kT} - 1} \cdot \left\{ 1 + \sum_{n=1}^{\infty} \frac{(8x^2 T^2 / T_0^2)^{2n}}{(2n+1)!(4n+1)!} \prod_{m=1}^{2n} \left[\left(\frac{\Delta U}{2\pi kT}\right)^2 + m^2 \right] \right\}. \quad (5)$$

$\Delta U = 4x^2 w \exp\left\{-\frac{4}{3} x^2 \sum_{s\lambda} |z_{s\lambda}|^2 (2\bar{n}_\lambda + 1)\right\}$ is the energy difference between the final and the initial state. We have also introduced the characteristic temperature T_0 , defined by the equation $(T/T_0)^2 = (8/3) \sum_{s\lambda} |z_{s\lambda}|^2 \bar{n}_\lambda$.

REFERENCE

- (1) Gosar P. and Kranjc T., Phonon Scattering in Solids, ed. by L.J.Challis, V.W.Rampton, and A.F.G.Wyatt, Plenum Publ. Corp., New York (1976), 217.