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TRAJECTORY TREATMENT OF THE $F_A^{(II)}$-CENTER IN $KCl:Li$. DECAY TIME IN THE EXCITED STATE

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Abstract - We simulate the temperature dependence of the decay time of an impurity in its excited state by obtaining the classical trajectories of the ions in the crystal. The $F_A^{(II)}$-center in $KCl:Li$ is treated.

I - INTRODUCTION

Recent developments of monitoring techniques in the range of picoseconds have allowed measurements of configurational relaxation times $T_R$ in excited states of impurities in crystals. By recording the rise time of emission from the relaxed excited state, a decrease in $T_R$ from about 15 ps to 1-2 ps was observed for the $F_A^{(II)}$ center in $KCl:Li$ upon raising the temperature to about 300K /1/ and about the same decrease was found more recently for the $^1T_{1u}$-state in $Kl:Ag$ /2/. The analyses of the data, which were able to account for only a small fraction of the energy dissipated in the configurational relaxation, employed a multiphonon relaxation formula (Eq. 3.36 in /3/) for the temperature dependence of $T_R$. This quantum mechanical expression gives the residence time $\tau_R$ in the initial state before transition to a neighboring level belonging to a different state.

We have performed computation of the classical trajectories of the ions for the $F_A^{(II)}$ center following excitation. The model for the center is shown in Figure 1, where the horizontal axis represents neighbor-correlated jumps of the vacancy between equivalent $100$ type positions. We use a crystal-mode representation (amplitude $Q_{q,s}$, mode index $s$, wave number $q$) for the ionic displacements, instead of the more common individual atom displacements. We solve about $10^4$ equations of motions for $Q_{q,s}$ and obtain the time of residence $\tau_R$ as well as decay ($\tau'$) and correlation ($\tau_{cor}$) for the decay of $Q_{vac}$.

The classical motions proceed along an adiabatic potential energy surface (APES) that is stable (namely, harmonic) for all displacements except $Q_{vac}$. In Figure 2 we show the relationship of our classical model with the multiphonon-relaxation model in /1/. Their formalism implies tunneling from the initial non-adiabatic $V_2$ to the surfaces $V_1$ or $V_3$. 

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Fig. 1 - Ionic configurations near the $F_{\text{A}}$(II) center, potentials in the ground and excited states and the experimental scheme of $/1/$ which monitored the emission rise-time.

Fig. 2 - Relation between multiphonon transition mechanism $V_2 \rightarrow V_3$ and classical trajectory on APES.
II - FORMALISM

The excited-state potential, having a saddle point at $Q_{q,s}=0$ and a decrease along $Q_{vac}$ in the 110 ($=x'$) direction, is $(k=q,j=s)$

$$V_{ex} = (1/2)\hbar\omega_k (k/k)^2 \sum_{j} (Q_k,j(t)-Q_{vac}=k,j(k/Nk) \rangle^{1/2} + (1/2)\Delta\cos\frac{\sqrt{2\pi}}{\alpha} Q_{vac}(t)$$

where

$$Q_{vac} = \frac{1}{\sqrt{N}} \sum_{k=1}^{K} \sum_{j=q}^{J} Q_k,j^{q,k,j} (k/k) \rangle^{1/2} \quad \text{and} \quad q^{q,s} = \frac{Q_{k'}}{q'} F(\omega_{q,s}), F(\omega) e^{-\omega/\omega_D},$$

$e_{q,s}$ and $F(\omega)$ give the mode selective in the composition of $Q_{vac}$; $\omega_D$ is the circular Debye frequency.

The initial conditions for the amplitudes at temperature $T$ are

$$Q_{q,s}(t=0) = \left(\frac{2k_BT_{q,s}}{m_{q,s}\omega_{q,s}}\right)^{1/2} \cos q_{q,s} \quad q_{q,s}(t=0) = \left(\frac{2k_BT_{q,s}}{M_{q,s}}\right)^{1/2} \sin q_{q,s}$$

where

$$k_B T^{-1} = (n_{q,s} + 1/2) \hbar \omega_{q,s} \quad n_{q,s} = (\exp(\hbar \omega_{q,s}/k_B T) - 1)^{-1} \quad \alpha \text{ are random angles.}$$

III - RESULTS

The amplitude $Q_{vac}$ shows three characteristic behaviors, depending on the energy excess $\Delta E$ and the Debye frequency $\nu_D = \omega_D/2\pi$. For aperiodic motion, in the lower portion, one associates the decay time with the time of stay $\tau_r$ or residence time near the $Q_{q,s}=0$, 100 position.

Quantitatively, this appears as in Figure 4, where curves are shown for three values of the high-frequency cut-off parameter $A$ in $F(\omega)$ and for simulations with a number of modes $\sim \frac{3}{4} K^3 \sim 10^4 (K=30)$.

![Fig. 3 - "Phase diagram" for the motion of $Q_{vac}$ in $\Delta E-\nu_D$ parameter space, $\xi = \frac{\Delta E}{\hbar \omega_D}$.](image-url)
Fig. 4 - The vacancy mode amplitude rises after a residence time of cca 2-5 Debye periods and performs a decaying oscillatory motion around the RES configuration.

Fig. 5 - Computed residence and correlation times as function of temperature
Raising the temperature lowers the residence time $\tau_r$ (Fig. 5). The decay of the damped oscillation represented by $\tau_{cor}$ in Figure 3 is likewise accelerated by a temperature rise. Results are given in /4,5/.

REFERENCES