Submitted to Physics Letters A

LBL-23
Preprint

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July 22, 1971

AEC Contract No. W-7405-eng-48

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TEMPERATURE DEPENDENCE OF DAMPING OF LATTICE RESONANCE IN ALKALI HALIDES*

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ABSTRACT

It is shown that the explicit T^2 dependence of damping due to quartic contributions is small in comparison with the T dependence from cubic anharmonicity. A two-time Green's function method is used.

* This work was supported in part by the U. S. Atomic Energy Commission.

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The problem of explaining the temperature dependence of the damping for lattice resonance of alkali halides has received considerable attention [1]. Recently, Mooij [2] pointed out that (1) previous theoretical considerations neglected the implicit temperature dependence of thermal expansion and (2) the quartic anharmonic contributions to the damping are negligible if thermal expansion is considered.

We have derived a general expression for the complex susceptibility tensor by use of two-time Green's functions. The susceptibility is a function of the frequency shifts $\Delta(\omega)$ and corresponding damping constant $\Gamma(\omega)$. The usual absorption coefficient is proportional to the imaginary part of the susceptibility tensor.

It is found that the cubic anharmonic contributions give rise to T $(n \to kT)$ dependence for the damping in the classical limit, and the quartic contributions introduce an additive T^2 dependence to the damping. However, the quartic contributions are of the order $O(1/N^2)$ compared to $O(1/N)$ for the cubic contributions.

The two-time retarded Green's functions used are defined by [3]

$$G(t,t') = -i\theta(t - t')\langle[A(t),B(t')]\rangle ,$$

$$= \langle\langle A; B\rangle\rangle = G(A; B), \qquad (1)$$

where

$$\theta(t - t') = \begin{cases} 1, t - t' > 0 \\ 0, t - t' < 0 \end{cases} , \qquad (2)$$

and the canonical ensemble average is denoted by

$$\langle \cdots \rangle = \frac{\mathrm{Tr}(e^{-\beta H}\cdots)}{\mathrm{Tr}\, e^{-\beta H}} ; \qquad \beta = \frac{1}{kT} . \tag{3}$$

The operators A and B are second quantized phonon operators in the Heisenberg representation. The Hamiltonian of the isolated system is denoted by H. The equation of motion of $G(t,t')$ is obtained by differentiating eq. (1) with respect to time. On taking the Fourier transform of this equation of motion, we obtain (in the energy representation)

$$EG(A;\, B)_E = \frac{1}{2\pi} \langle [A,B] \rangle_E + G([A,H];\, B)_E , \tag{4}$$

where $E = \omega + i\epsilon$ and $\hbar = 1$. The frequency of the applied field is given by ω.

It is well known that the linear complex susceptibility is given by [3]

$$\chi_{\mu\nu}(\omega) = -2\pi\, G(M_\mu;\, M_\nu)_E , \tag{5}$$

where M is the electric moment of the crystal. The directions of the applied electric field and the response are ν and μ respectively. The Hamiltonian and electric moment in terms of phonon creation and annihilation operators are [4]

$$\begin{aligned} H = H^+ &= H_0 + H_3 + H_4 , \\ &= \sum_{\underset{\sim}{\alpha}} \omega_{\underset{\sim}{\alpha}}\, a^+_{\underset{\sim}{\alpha}}\, a_{\underset{\sim}{\alpha}} + \sum_{\underset{\sim}{\alpha},\underset{\sim}{\beta},\underset{\sim}{\gamma}} V^{(3)}_{\underset{\sim}{\alpha},\underset{\sim}{\beta},\underset{\sim}{\gamma}}\, A_{\underset{\sim}{\alpha}}\, A_{\underset{\sim}{\beta}}\, A_{\underset{\sim}{\gamma}} \\ &\quad + \sum_{\underset{\sim}{\alpha},\underset{\sim}{\beta},\underset{\sim}{\gamma},\underset{\sim}{\rho}} V^{(4)}_{\underset{\sim}{\alpha},\underset{\sim}{\beta},\underset{\sim}{\gamma},\underset{\sim}{\rho}}\, A_{\underset{\sim}{\alpha}}\, A_{\underset{\sim}{\beta}}\, A_{\underset{\sim}{\gamma}}\, A_{\underset{\sim}{\rho}} , \end{aligned} \tag{6}$$

and

$$M_\mu = N^{\frac{1}{2}} \sum_{\underset{\sim}{0}} \frac{M_\mu(\underset{\sim}{0})}{(2\omega_{\underset{\sim}{0}})^{\frac{1}{2}}} A_{\underset{\sim}{0}} , \tag{7}$$

where $A_{\underset{\sim}{\alpha}} = a^+_{-\underset{\sim}{\alpha}} + a_{\underset{\sim}{\alpha}}$ and $A_{\underset{\sim}{0}} = a^+_{\underset{\sim}{0}} + a_{\underset{\sim}{0}}$. The indices $\underset{\sim}{\alpha}$, $\underset{\sim}{\beta}$, $\underset{\sim}{\gamma}$, and $\underset{\sim}{\rho}$ represent both the wave vector $\underset{\sim}{k}$ and polarization index j. For example, $\underset{\sim}{\alpha} = \underset{\sim}{k}j$ and $\underset{\sim}{\beta} = \underset{\sim}{k}'j'$. The expansion coefficients in eqs. (6) and (7) are discussed by Born and Huang [5].

On substituting eq. (7) into eq. (5), we obtain

$$\chi^{(\omega)}_{\mu\nu} = -2\pi \frac{N}{2} \sum_{\underset{\sim}{0},\underset{\sim}{0}'} \frac{M_\mu(\underset{\sim}{0})\, M_\nu(\underset{\sim}{0}')}{(\omega_{\underset{\sim}{0}}\omega'_{\underset{\sim}{0}})^{\frac{1}{2}}} [G^1 + G^2] , \tag{8}$$

where

$$G^1 = G(a^+_{\underset{\sim}{0}};\, A'_{\underset{\sim}{0}})_E \tag{9}$$

and

$$G^2 = G(a_{\underset{\sim}{0}};\, A'_{\underset{\sim}{0}})_E . \tag{10}$$

The scheme for solving the system of dependent equations is

$$G(1;\, \cdots) \rightarrow (\text{constant})_1 + f_1(\omega)\, G(1;\, \cdots) + g_2(2;\, \cdots) + g_3(3;\, \cdots), \tag{11}$$

$$\begin{aligned} g_2(2;\, \cdots) \rightarrow\ & \underset{\substack{\| \\ 0}}{(\text{constant})_2} + f_2(\omega)\, g_2(2;\, \cdots) \\ & + \Gamma_3(3;\, \cdots) + \Gamma_4(4;\, \cdots), \\ & \downarrow \leftarrow \text{decoupling} \rightarrow \downarrow \\ & \gamma^{(3)}(E,n)\, G(1;\, \cdots) + \xi^{(1)}(n)\, g_2(2;\, \cdots) \end{aligned} \tag{12}$$

$$g_3(3;\cdots) \rightarrow (\text{constant})_3 + f_3(\omega)\, g_3(3;\cdots) + \Gamma_4(4;\cdots) + \Gamma_5(5;\cdots) \quad , \tag{13}$$

$$\xi^{(2)}(n)\, g_2(2;\cdots) + \gamma^{(4)}(E,n^2)\, G(1;\cdots)$$

$\leftarrow$ decoupling $\rightarrow$

where $g_2(2;\cdots)$ and $g_3(3;\cdots)$ are higher-order Green's functions[2]. The numbers on the left of the semicolons in the Green's functions indicate the number of phonon operators present. The second, third, and fourth terms in each of the equations represent contributions from H_0, H_3, and H_4 respectively. The functions $\gamma^{(3)}(E,n)$ and $\gamma^{(4)}(E,n^2)$ are the cubic and quartic contributions to the damping; they give rise to T and T^2 dependence of the damping constant. However, the T dependence is $O(1/N)$ and the T^2 dependence is $O(1/N^2)$. The decoupling scheme is as follows

$$g_3^{\,1}(a^+_{-\underset{\sim}{\alpha}}\, a_{\underset{\sim}{\beta}}\, a^+_{-\underset{\sim}{\gamma}};\, A'_{\underset{\sim}{0}}) \approx [n_{\underset{\sim}{\beta}}\, \delta_{-\underset{\sim}{\alpha},\underset{\sim}{\beta}}\, \delta_{-\underset{\sim}{\gamma},\underset{\sim}{0}} + (n_{\underset{\sim}{\beta}} + 1)\, \delta_{\underset{\sim}{\beta},-\underset{\sim}{\gamma}}\, \delta_{-\underset{\sim}{\alpha},\underset{\sim}{0}}]G^1, \tag{14}$$

where off-diagonal terms are neglected and $n_{\cdots}$ is the usual average boson occupation number. A similar procedure is used to decouple the $\Gamma_5(5;\cdots)$ Green's functions giving rise to n^2 (or T^2) dependence in the classical limit.

The details of this work will be presented elsewhere.

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