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Ultrasonic Resonance and Relaxation in RbCl:OH

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Abstract. The change of elastic constants and mechanical loss from [100] elastic dipoles are calculated on the basis of a six level tunneling model. Resonance and relaxation mechanisms are treated together, and expressions for C' - and C_{11} - sound modes are given. The results are used to analyse experimental data for the RbCl:OH system. The low temperature ultrasonic attenuation is attributed to the resonance mechanism, and the frequency and the concentration dependence are interpreted in terms of dynamical and static interaction between dipoles.

1. INTRODUCTION

Low temperature mechanical and thermal properties of alkali halide crystals containing hydroxyl ions (OH^-) show anomalous behavior due to quantum mechanical tunneling [1]. The resonance absorption of sound wave in glasses has been studied extensively by means of a phenomenological two level system (TLS) model [2]. The OH^- ions in alkali halide crystals could be a model system for the TLS. The OH^- ions lie on [100] crystal axes and the elastic response could be treated quantitatively by the elastic dipole model. We have developed a full expression for the change of elastic constants due to the elastic dipole. The results are applied to the analysis of our experimental results on RbCl:OH [3].

2. CALCULATION OF ELASTIC CONSTANTS

A [100] elastic dipole has a tetragonal strain and interacts with external tetragonal stress components. The origin of external stress may be an applied uniaxial stress or internal stress. We choose an xyz -coordinate system along the cubic axes. There are 6 allowed states, in which the OH^- ion is directed to $x, -x, y, -y, z, -z$ orientations. The Hamiltonian for the dipole under the static external stress can be written as

$$H^0 = H^L + H^T + H^S. \quad (1)$$

Where H^L is the lattice hamiltonian and the eigen functions are $\psi_x, \psi_{-x}, \psi_y, \psi_{-y}, \psi_z, \psi_{-z}$. If there are no external stresses (or strains), these six states are equivalent and should have same energies. We may choose the energy to be zero and use suffixes i or j to express these states in general expressions. H^T is the tunneling hamiltonian and H^S corresponds to the interaction between the dipole and the external stress. The interaction energy for the dipole along the x, y, z axis can be expressed as

$$\begin{aligned} S_x &= -v_0 \delta \lambda (2\sigma_{xx} - \sigma_{yy} - \sigma_{zz})/3, \\ S_y &= -v_0 \delta \lambda (2\sigma_{yy} - \sigma_{zz} - \sigma_{xx})/3, \\ S_z &= -v_0 \delta \lambda (2\sigma_{zz} - \sigma_{xx} - \sigma_{yy})/3, \end{aligned} \quad (2)$$

where v_0 is the volume occupied by a dipole and $\delta \lambda = \lambda_1 - \lambda_2$. λ_1 and λ_2 are ellipsoidal strain components produced by a dipole. Then the matrix elements of H^0 are written as

$$H_{ij}^0 = -\frac{\Delta}{2} \begin{bmatrix} 0 & 0 & 1 & 1 & 1 & 1 \\ 0 & 0 & 1 & 1 & 1 & 1 \\ 1 & 1 & 0 & 0 & 1 & 1 \\ 1 & 1 & 0 & 0 & 1 & 1 \\ 1 & 1 & 1 & 1 & 0 & 0 \\ 1 & 1 & 1 & 1 & 0 & 0 \end{bmatrix} + \begin{bmatrix} S_x & 0 & 0 & 0 & 0 & 0 \\ 0 & S_x & 0 & 0 & 0 & 0 \\ 0 & 0 & S_y & 0 & 0 & 0 \\ 0 & 0 & 0 & S_y & 0 & 0 \\ 0 & 0 & 0 & 0 & S_z & 0 \\ 0 & 0 & 0 & 0 & 0 & S_z \end{bmatrix} \quad (3)$$

Here, Δ is the tunneling energy. Six eigen states Ψ_m and energies E_m for the dipole under the external stress are determined from the Schrödinger equation

$$H^0 \Psi_m = E_m \Psi_m \quad (4)$$

by diagonalizing the hamiltonian H^0 . Ψ_m is expanded by ψ_i as

$$\Psi_m = \sum_i a_{mi} \psi_i. \quad (5)$$

There are six eigen states E_m , three of them ($m = 1-3$) are the solutions of a cubic equation

$$E^3 + (S_x S_y + S_y S_z + S_z S_x - 3\Delta^2) + 2\Delta^3 - S_x S_y S_z = 0, \quad (6)$$

and the others are

$$E_4 = S_x, \quad E_5 = S_y, \quad E_6 = S_z. \quad (7)$$

As an example, these energy branches under the stress ($\sigma_{xx} = 0, \sigma_{yy} = \sigma, \sigma_{zz} = -\sigma$) are shown in Figure 1, where the branches shown by dashed lines are inactive to the sound wave.

The matrix elements of the hamiltonian for the interaction between sound wave and elastic dipole, H'_{ij} is similar to H^S .

$$H'_{ij} = h_{ij} \delta_{ij} \sin(\omega t), \quad (8)$$

where ω is the angular frequency of the sound wave. The matrix elements h_{ij} depend on the mode of the sound wave. Two types of the pure sound modes, C_{11} - and $2C' (= C_{11} - C_{12})$ -mode, interact with the $[100]$ dipole. The strain component due to C_{11} -mode propagating along the x -direction is $\epsilon_{xx} = \epsilon \sin(\omega t)$ and

$$h_{xx} = -2q, \quad h_{yy} = q, \quad h_{zz} = q; \quad q = (C_{11} - C_{12})v_0(\delta\lambda)\epsilon/3. \quad (9)$$

The strain for the C' -mode is given by $\epsilon_{xx} = -\epsilon_{yy} = \epsilon/2$, and the matrix elements are

$$h_{xx} = -q', \quad h_{yy} = q', \quad h_{zz} = 0; \quad q' = (C_{11} - C_{12})v_0(\delta\lambda)\epsilon/2. \quad (10)$$

We proceed to calculate the response of the dipole to the sound wave by time dependent perturbation theory. The excess strain produced by dipoles is expressed as

$$\Delta\epsilon_{ii} = -\frac{2}{3} \frac{n_0 v_0 \delta \lambda}{k_B T} \sum_m E_m \sum_{n(\neq m)} f_{mn} a_{mi} a_{ni} + 2v_0 \delta \lambda \sum_m n_m(t) a_{mi}^2. \quad (11)$$

Where,

$$n_m(t) = -\frac{n_0}{6k_B T} b_{mm} \left[\frac{\sin \omega t}{1 + (\omega \tau_1)^2} + \frac{\cos \omega t}{1 + (\omega \tau_1)^2} \right], \quad (12)$$

$$f_{mn} = -\frac{b_{mn}}{2\hbar} [X_{mn}(\omega) \sin \omega t + Y_{mn}(\omega) \cos \omega t], \quad (13)$$

$$X_{mn}(\omega) = \frac{(\omega_{mn} - \omega)\tau_2^2}{(\omega_{mn} - \omega)\tau_2^2 + 1} + \frac{(\omega_{mn} + \omega)\tau_2^2}{(\omega_{mn} + \omega)^2\tau_2^2 + 1}, \quad (14)$$

$$Y_{mn}(\omega) = \frac{\tau_2}{(\omega_{mn} - \omega)\tau_2^2 + 1} + \frac{\tau_2}{(\omega_{mn} + \omega)^2\tau_2^2 + 1}, \quad (15)$$

$$\omega_{mn} = (E_m - E_n)/\hbar, \quad b_{mn} = \sum_i a_{mi} a_{ni} h_{ii}. \quad (16)$$

Figure 1: Energy levels under C' stress.

Figure 2: Resonance absorption

a) $\omega\tau_2 = 10$: b) $\omega\tau_2 = 1$.

Where τ_1 is the relaxation time and τ_2 is the life time of the dipole. These have similar meanings to the longitudinal and the transverse relaxation time in the theory of magnetic resonance. The first term on the RHS of eq.(11) corresponds to the resonance interaction of the dipoles with the sound wave. The second term corresponds to the relaxation of the dipoles through the change of population n_m in the level m . The change of elastic constants $\Delta C_{11}/C_{11}$ and $\Delta C'/C'$ are given by

$$\Delta C_{11}/C_{11} = \Delta\epsilon_{11}/\epsilon_{11}, \quad \Delta C'/C' = (\Delta\epsilon_{11} - \Delta\epsilon_{12})/(\epsilon_{11} - \epsilon_{12}). \quad (17)$$

As we have assumed the incident sound to be $\epsilon \sin \omega t$, the in-phase component corresponds to the change of sound velocity, and the out-of-phase component corresponds to the absorption or mechanical loss.

As an example, the loss angles (δ/π) for the resonance absorption under the internal stress S are shown in Figure 2, where labels a, b, and c represents the sound frequency $\omega = 0.5, 1.0$ and $1.5\Delta/\hbar$. When the dipoles have a long life time ($\omega\tau_2 = 10$), sharp resonances happen as the energy gap matches to frequency. On the other hand the peak becomes broader when τ_2 is small.

3. RESONANCE AND RELAXATION IN RbCl:OH

Ultrasonic attenuation and velocity in RbCl:OH system have been measured by the present authors [3]. A typical result is shown in Figure 3. A relaxation peak and the velocity dispersion are seen at around 6 K. Below 3 K the attenuation increases with decreasing temperatures due to the resonance. The temperature dependence is fitted to the expression

$$\delta/\pi = A + \Delta_{da}/T. \quad (18)$$

The value Δ_{da} represents the strength of the resonance absorption. The frequency dependence of Δ_{da} for three specimens (9, 15, and 57 ppm OH) are summarized in Figure 4. It seen that the frequency dependence is negative ($\propto -0.45$) for the lower concentration specimens and positive ($\propto .36$) for the high concentration specimen. This qualitative difference of the resonance absorption may be attributed to the difference of life time τ_2 . Hess et al. have measured the dielectric absorption in RbCl:OH [4], and found that τ_2 strongly depends on the OH concentration due to the interaction between dipoles. τ_2 in a 100 ppm specimen was found to be three orders of magnitude smaller than in a 9 ppm specimen.

As shown in Figure 2, resonance absorption shows sharp peaks when $\omega\tau_2 \gg 1$, then it is sensitive to the distribution of internal stress. The observed negative frequency dependence is possible when the number of dipoles is smaller for higher internal stress. The situation is reasonable if the origins

Figure 3: Ultrasonic attenuation and velocity change in RbCl:OH.

Figure 4: Measured resonance strength Δ_{da} .

of the internal stress are point defects or dislocations. In the high concentration specimen (57 ppm OH) τ_2 is smaller and the resonance peak becomes broader, then the strength of absorption is less sensitive to the stress distribution and a positive frequency dependence is realized.

The relaxation peak and the velocity dispersion observed at higher temperature are related to the relaxation time τ_1 . From the analysis of both data, the temperature dependence of τ_1 has been determined. The derived τ_1 is proportional to T^{-1} and T^{-4} below and above 4 K, respectively. This result suggests that the relaxation is not due to a thermal activation process. T^{-1} and T^{-4} dependences are considered to be due to the one-phonon and the multi-phonon processes.

4. CONCLUSION

The resonance and the relaxation phenomena of [100] elastic dipole are treated systematically on the basis of a quantum mechanical tunneling model. RbCl:OH may be a suitable system to test the theory, because it has a proper relaxation time and both phenomena can be observed at different temperatures in a specimen. The frequency and the concentration dependences of the resonance absorption reveal the static interaction between the dipole and the internal stress, and the dynamical dipole-dipole interaction. Similar phenomena are observed in magnetic systems. The energy diagram for the elastic dipole could be determined by scanning the external stress and observing the resonance absorption.

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