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Study of quasielastic light scattering in crystals

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In this doctoral thesis, the origin of the quasielastic light scattering consisting of two components in crystals, which has been subject for over twenty years in this field has been clarified both experimentally and theoretically through consideration of two-phonon difference Raman process instead of the conventional thermodynamic and hydrodynamic theory.

Light scattering experiments were performed for several kinds of crystals, including rutile (TiO_2), ZnSe, Si, and $SrTiO_3$, which were found to exhibit quasielastic light scattering consisting of two components, nameley, the "narrow component" and the "broad component". Temperature dependences of the linewidth and that of the intensity of both the narrow and the broad quasielastic components were analyzed, and it was found that the conventional macroscopic light scattering theory, which accounts for the narrow component in the high temperature region cannot explain the whole features of the observed two quasielastic components. We then formulated instead an explanation based on two-phonon difference Raman processes for both components.

The narrow component is well explained by two-phonon difference processes from a single acoustic phonon branch. With this model, the temperature dependence of the integrated intensity was consistent with our observations, and the temperature induced line narrowing was reproduced qualitatively (Fig.1). At high temperatures, however, the narrow component may be better explained by conventional entropy fluctuations since the observed linewidth approaches the values expected from thermal diffusion theory. The wave-vector (Q) dependence of the narrow component differs in each scattering mechanism, i.e., the linewidth is proportional to Q in light scattering due to two-phonon difference processes from a single phonon branch, while it is proportional to Q^2 in light scattering by entropy fluctuations. We found, however, that the wave-vector dependence of the narrow component linewidth does change from Q^2 to Q with decreasing temperature. It is concluded that this change in wave-vector dependence is due to infrequent phonon collision in the low temperature region causing break-down of hydrodynamic and thermodynamic description in such a low temperature region.

Macroscopic light scattering theory cannot also account for the broad quasielastic component. We have shown that two-phonon difference Raman processes from *different* phonon branches near zone boundary provide a good explanation for the temperature dependent intensity of the broad quasielastic component. Furthermore, temperature dependence of the linewidth and its insensitivity to changes in scattering wave vector are also explained with this model.

Finally, we have shown that two kinds of quasielastic components which have entirely different characteristics were uniformly explained by two-phonon difference Raman process which is a universal scattering mechanism in crystals. This suggests a possibility that quasielastic light scattering essentially consists of two components in any crystals.

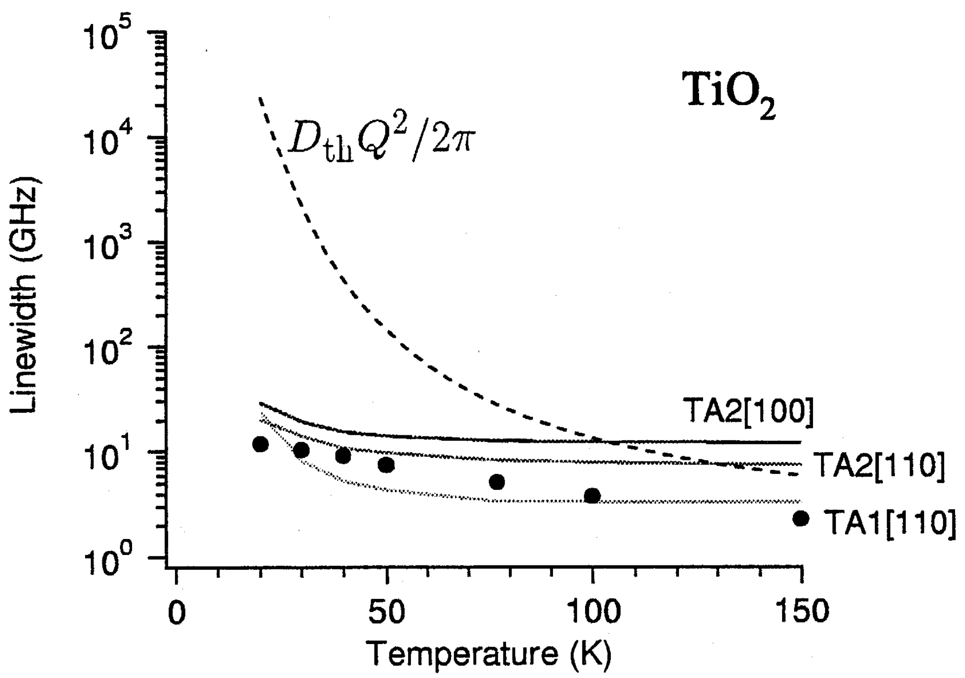

Fig. 1: Temperature dependence of the narrow component linewidth in rutile (TiO_2). The filled circles, the dashed line, and the labeled solid lines are experimentally measured linewidth, linewidth calculated from conventional macroscopic theory (D_{th} is thermal diffusivity), and linewidths calculated from two-phonon difference processes from a single phonon branch (labels indicate the corresponding phonon modes), respectively. It is to be noted that the last three lines were obtained without any adjustable parameters.