

Two-phonon bound states in imperfect crystals*

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Abstract. The question of the occurrence of two-phonon bound states in imperfect crystals is investigated. It is shown that the anharmonicity mediated two-phonon bound state which is present in perfect crystals gets modified due to the presence of impurities. Moreover, the possibility of the occurrence of a purely impurity mediated two-phonon bound state is demonstrated. The bound state frequencies are calculated using the simple Einstein oscillator model for the host phonons. The two-phonon density of states for the imperfect crystal thus obtained has peaks at the combination and difference frequencies of two host phonons besides the peaks at the bound state frequencies. For a perfect crystal the theory predicts a single peak at the two-phonon bound state frequency in conformity with experimental observations and other theoretical calculations. Experimental data on the two-phonon infrared absorption and Raman scattering from mixed crystals of $GA_{1-c}Al_cP$ and $Ge_{1-c}Si_c$ are analysed to provide evidence in support of impurity-mediated two-phonon bound states. The relevance of the zero frequency (difference spectrum) peak to the central peak observed in structural phase transitions, is conjectured;

Keywords. Two-phonon bound states; imperfect crystals; anharmonic crystals. phonon-phonon interactions; combination modes.

1. Introduction

The existence of a sharp peak in the two-phonon Raman spectrum of diamond above twice the maximum single phonon frequency; first observed by Krishnan (1946) has subsequently been confirmed by Solin and Ramdas (1970). This peak was interpreted by Cohen and Ruvalds (1969) as a two-phonon bound state brought about by the quartic anharmonic interaction. Since then two-phonon bound states have become a subject of intense theoretical and experimental investigation. Ruvalds and Zawadowski (1970) from a Green's function calculation successfully demonstrated that two-phonon resonant and bound states could exist provided that the quartic anharmonic coupling constant is large. They also suggested that the effect of the two-phonon state could show up through interference in a single phonon spectrum. Maradudin (1971) solved for the two-phonon Green's function using the Fredholm method, in the presence of both the quartic and cubic anharmonic interactions and argued from symmetry considerations that the cubic term does not really contribute to the bound state for Bravais crystals. Using the Peierl's approximation for the quartic anharmonic coefficient he demonstrated that the strength of the coupling becomes too small to give a two-phonon bound state for germanium. On the contrary a similar calculation by Wu and Birman (1975) predicted the possibility for a

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