LOW - TEMPERATURE INTERFACE SPECIFIC HEAT DUE TO LOCALIZED MODES

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Green functions are derived for the displacement associated with acoustic vibrations in isotropic elastic media, and used to evaluate the surface specific heat in the harmonic approximation. The contribution of surface elastic waves of the Rayleigh and love types are pointed out and their features discussed. The nature of the results and their relations to previous works in this field is also presented and discussed.

1. INTRODUCTION

In this paper we present a Green function calculation of the surface contribution to the specific heat of a crystal. We restrict our calculation to the low - temperature limit case since, provided $k_B T/\hbar$ is very small, we can replace the dispersion relation for the three acoustic branches by its long wavelengths form.

The majority of the previous calculations on this subject were carried out using a lattice - dynamical calculation [1 - 4] together with Born cyclic boundary conditions. This can be justified by the relation between the vibrational spectrum and the propagation of disturbances through the crystal lattice (5), although (3) and (4) also used a Green function method to find the difference between the specific heat of the out and the perfect uncut crystal.

On the other hand, if one considers the Green function $\langle\langle u^\alpha(\vec{r}); u^\alpha(\vec{r}')^* \rangle\rangle$ as the evolution matrix or propagator for the system, a great deal of the surface system dynamics is very neatly contained in it. Burt (6) used this technique to set up a very useful method to calculate the surface specific heat since the density in frequency of the normal modes of a crystal is equal to the trace of the Fourier transform $\langle\langle u^\alpha(\vec{r}); u^\alpha(\vec{r}')^* \rangle\rangle_\omega$.

The interface properties of the crystals under consideration were already widely studied (7). The localized waves which can exist and propagate along the two crystalline interfaces considered here are known as Rayleigh and love waves. The calculation of the

necessary interface Green functions were done using linear response theory as described by Barker and Loudon (8).

We now describe our method briefly. Consider the effect of an externally applied point force

$$(F^\alpha/\bar{A})\exp(-i\omega t)\delta(\alpha - d) \quad (1)$$

where $\delta(\alpha - \alpha')$ is the Dirac delta function and $\alpha(\alpha')$ can be x, y or z (x' , y' , or z'). $\bar{A}$ is the surface area of the crystal. The energy of interaction of the force with the entire elastic crystal has the form (9).

$$H_{int} = - u^\alpha F^\alpha \exp(-i\omega t) \quad (2)$$

where u^α is the α - component of the displacement of the acoustic phonon. The form of (2) is required for the standard application of linear response theory (10).

The applied force (1) generalizes the elastic equation of motion in the interior of the crystal and, in view of the standard form (2) of the interaction energy, the required displacement Green function is simply equal to (9)

$$\langle\langle u^\alpha(\vec{r}); u^\alpha(\vec{r}')^* \rangle\rangle_\omega = u^\alpha(\vec{r})/F^\alpha \quad (3)$$

The specific heat in the harmonic approximation can be given by

$$C(T) = \int F(\omega) D(\omega) d\omega \quad (4)$$

where $F(\omega)$ is the specific heat of an harmonic oscillator of frequency ω and $D(\omega)$ is the density of modes. This density of modes can be obtained from the interfaces Green functions according to

$$D(\omega) = -\frac{2}{\pi} \text{Im} \left[\omega \int_{\alpha} \rho d^3r \langle\langle u^\alpha(\vec{r}); u^\alpha(\vec{r}')^* \rangle\rangle_\omega \right] \quad (5)$$

where ρ is the mass density of the crystal. Using (4) and (5) with the interface Green functions calculated from (3) we can then obtain the localized modes contribution to the surface specific heat of the crystals.

In section 2 of this paper we calculate the contribution of the Rayleigh waves to the interface specific heat while section 3 is devoted to the calculation of the love waves contribution. The conclusions are in section 4.

2. RAYLEIGH WAVE CONTRIBUTION

Consider an isotropic material occupying the half-space $z < 0$ with a flat surface, with area $\bar{A}$, in the plane $z = 0$ and vacuum in the half-space $z > 0$. Due to translational invariance, $\langle\langle u^\alpha(\vec{r}); u^\alpha(\vec{r}')^* \rangle\rangle_\omega$ is independent of the components of $\vec{r}$ parallel to the surface. Therefore (5) is reduced to

$$D(\omega) = -\frac{2\bar{A}\rho\omega}{\pi} \left[\text{Im} \int dz \langle\langle u^z(z); u^z(z')^* \rangle\rangle_\omega \right] \quad (6)$$

For modes polarized in the plane of propagation there is a frequency region where ω is below the threshold for transverse waves, i.e. $\omega < \omega(q)$. Only surface wave of the Rayleigh type can propagate in this region obeying the dispersion relation

$$4V_T^3(V_L^2 - V_R^2)^{1/2}(V_T^2 - V_R^2)^{1/2} = V_L(2V_T^2 - V_R^2)^2 \quad (7)$$

In (7), V_T and V_L are respectively the transverse and longitudinal velocity of the acoustic wave in the semi-infinite crystal; V_R is the velocity of the surface Rayleigh wave.

The contribution of this localized mode to the interface specific heat of the crystal is now calculated from (4) and (6); the Green functions used in this calculation (11) take into account only surface contributions. The result is

$$C(T) = 12\pi K_B \zeta(3) (K_B T/h V_R)^2 \quad (8)$$

in agreement with Burt (6). Here $\zeta(x)$ is the Riemann Zeta function K_B and h are the Boltzmann and Planck constant respectively. If we consider, for instance, the isotropic half-space as a sample of Al crystal we can find that at $T = 300^\circ\text{K}$, $C \approx 2.95 \times 10^{-3} \text{ J/Km}^2$.

3. LOVE WAVE CONTRIBUTION

Consider a film of thickness H deposited on a substrate whose thickness H' is effectively infinite. The film and substrate are made from different materials, both optically and elastically isotropic. For modes with polarization perpendicular to the plane of propagation there is a frequency region given by

$$vq^x < \omega < v'q^x \quad (9)$$

where the system supports the propagation of another kind of surface wave, namely the love waves. In (6), $v(v')$ is the transversal velocity of the acoustic wave of wave-vector $q(q')$ in the film (substrate). The dispersion relation of this wave is (12)

$$C^{44} q^z \tan q^z H = C'^{44} \gamma \quad (10)$$

where C^{44} and C'^{44} are components of the elastic tensor in the film and substrate respectively, and

$$q'^z = i\gamma = \left[(\omega/v')^2 - (q^x)^2 \right]^{1/2} \quad (11)$$

with similar expression for q^z .

In order to see some properties of the love waves it is convenient to re-express the love wave condition (10) as

$$\tan q^z H = \frac{C^{44} \left[\beta^2 \omega^2 - (q^z)^2 \right]^{1/2}}{C^{44} q^z} \quad (12)$$

where

$$\beta^2 = (v'^2 - v^2)/v^2 v'^2 \quad (13)$$

In reference to (12) we have that in general there are n waves for $(n-1)\pi/H < \beta\omega < n\pi/H$. Also, the n th love wave starts at the point $q^x = (n-1)\pi v/(\beta v'^2 - v^2)^{1/2} H$ and $\omega = v'q^x$; it has the asymptotic behaviour $\omega \approx v'n^x$ for large ω .

The density of states is now

$$D(\omega) = \frac{1}{2\pi\omega^2 N} \left(\frac{q^2}{q^z} B_1 + \frac{q'^2}{2\gamma^2} B_2 \right) \sum_n \delta(\omega - \omega_n) \quad (14)$$

where

$$N = \frac{1}{v^2 q^z} \left(C^{44} + C'^{44} \gamma H \right) \sin q^z H + \left(\frac{C'^{44}}{v'^2 \gamma} + \frac{C^{44}}{v^2} \right) \cos q^z H \quad (15)$$

$$B_1 = (C^{44} + C'^{44} \gamma H) \sin q^z H + C^{44} q^z H \cos q^z H \quad (16)$$

$$B_2 = C^{44} q^z \sin q^z H + C'^{44} \gamma \cos q^z H \quad (17)$$

and the summation in n takes into account all the love wave modes. The required love wave contribution to the surface specific heat can be easily calculated from (14) and (4) to give

$$C(T) = 6 K_B \left(\frac{K_B T}{h q^x} \right)^2 \zeta(3) \sum_n \frac{1}{(v_L^L)^4} \quad (18)$$

where

$$I = \frac{1}{N} \left(\frac{q^2}{q^z} B_1 + \frac{q'^2}{2\gamma^2} B_2 \right) \quad (19)$$

and v_n^L is the velocity of the n th love wave mode. For the case of a film of amorphous SiO_2 on an amorphous MgO substrate, using the materials parameters as give in (12), we can estimate at $T = 300^\circ\text{K}$, $C \approx 1.13 \times 10^{-4} \text{ J/Km}^2$, one order of magnitude less than the localized Rayleigh mode contribution to the surface specific heat.

4 CONCLUSIONS

The main purpose of this work is a theoretical prediction about the contributions to the surface specific heat of isotropic elastic solids due to localized modes of the Rayleigh and Love types. For the Rayleigh mode the calculation, using the method developed here, is quite straightforward provided one knows the appropriate interface Green functions, although the same does not occur for the Love modes. The main difficulty that arises with these surface modes concerns the fact that as q^x increases the number of modes with the same ω but different q^x also increases. However, by an adequate choice of the parameters β and H , we can restrict our calculations to a fixed number of modes and even, as we are in the low-temperature regime, to

consider only one mode in the calculations as it is illustrated in ref. (12).

A more realistic calculation should include elastic anisotropic effects. The inclusion of these effects in the elastic equation of motion however, does not affect too much our results since this anisotropy can be incorporated into the components of the elastic tensor.

We can justify the harmonic approximation used here because it has the great advantage of simplifying the evaluation of the required Green functions, otherwise very hard to be found.

Finally we would like to remark that the present method may be considered as an extension of previous theoretical works in surface specific heat, as far as localized modes are concerned.

REFERENCES

1. M DUPUIS, R. Mazo, and L. Onsager, J. Chem. Phys. 33, 1452 (1960)
2. R. STRATTON, J. Chem. Phys. 37, 2972 (1962)
3. A.A. MARADUDIN and R.F. Wallis, Phys. Rev. 148, 945 (1966)
4. B. DJARFARI - Rounani and L. Dobrzynski, Phys. Rev. 14, 2296 (1976)
5. M. BORN and K. Huang, Dynamical Theory of Crystal Lattices, Clarendon Press, Oxford (1968)
6. M.G. BURT, J. Phys. C 6, 855 (1973).
7. B.A. AULD, Acoustic Fields and Waves in Solids, Vols 1 and 2, Wiley, N.Y. (1973)
8. A.S. BARKER Jr, and R. Loudon, Rev. Mod. Phys. 44, 18 (1972)
9. R. LOUDON and J.R. Sandercock, J. Phys. C 13, 2609 (1980)
10. L.D. LANDAU and E.M. Lifshitz, Statistical Physics, Pergamon Press, Oxford (1968)
11. R. LOUDON, J. Phys. C 11, 2623 (1978)
12. E.L. ALBUQUERQUE, R. Loudon and D.R. Tilley, J. Phys. C 13, 1775 (1980)