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# Phonon localization in two-dimensional self-supported films

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Phonon localization in thin self-supported films is investigated. The localization in the sense of Anderson depends critically on inelastic phonon processes; defects and roughness at the film boundaries are assumed to result in resonant phonon scattering by two-level systems. We define a phonon localization length $\xi$ from the elastic diffusion determined by two-dimensional isotope scattering and by partially specular reflection from boundaries. The localization frequency edge $\omega_0$ is shown to depend essentially on the conditions of the film surfaces.

## I. INTRODUCTION

Since its discovery by Abrahams *et al.*,[1] the problem of weak localization of electrons in a disordered medium has received considerable attention both theoretically and experimentally.[2] The localization in the sense of Anderson[3] is a wave phenomenon corresponding to an interference of electrons scattered at impurities;[4] therefore a similar effect can be expected to occur for phonons in solids.[5] During the past decade much theoretical work has been devoted to the study of phonon localization in solids;[5–12] either in two- or three-dimensional solids, generally with mass disorder.[7,8] The localization effects being stronger in one-dimensional systems, phonon localization in one-dimensional solids has been studied extensively[13,14] in the past; more recently an acoustic Anderson localization experiment has been done by He and Maynard.[15]

As was first pointed out by Kelly,[16] the observation of phonon localization in solids requires free-standing samples of low dimensionality.[4] Indeed, self-supporting films or wires are necessary to study the intrinsic thermal conductance; any structure on a substrate will undergo coupling between the phonons in the structure and the bulk three-dimensional phonons in the supporting substrate. Free-standing films or wires are now rather easily prepared by lithography techniques.[17] In wires of small cross section and in films of very small thickness, the phonon transport is, respectively, either one or two dimensional at low temperatures because of the size quantization.[18,19] This leads to restricted phase spaces and therefore to most favorable cases of interference phenomena over long phase coherence times.

The phonon localization depends on the relative magnitudes of the inelastic scattering rate $\tau_i^{-1}(\omega)$ and the elastic scattering rate $\tau_e^{-1}(\omega)$ that are both frequency dependent: the inelastic scattering rate must be smaller than the elastic scattering rate leading to localization in order that a phonon wave packet can travel across a region of localization without being transferred to another wave packet of localized states.

The question we wish to address in this paper is the possibility to observe phonon localization in two-dimensional dielectric films at low temperatures ($\sim 1$ K). We consider here thin free films of thickness $d = 100-200$ Å, length $L$, and width $w$. In such small thickness films, the phonon spectrum exhibits size quantization,[18] since at 1 K, the thermal phonon wavelength $\lambda_{\mathrm{th}} \sim 800$ Å $\gg d$; the lattice vibrational modes are confined in their propagation along the film thickness. We are then left with two-dimensional phonon transport with thermal conductance along the length $L$ of the film.

## II. SURFACE EFFECTS

In contrast to ideal acoustic surfaces, the lateral surfaces of the film considered here are real surfaces with defects and roughness that destroy the translational symmetry of the surface. It is known that real surfaces are rough on the scale $\delta$ of hundreds of Å,[20] i.e., of the order of magnitude of the mean thermal phonon wavelength $\lambda_{\mathrm{th}}$: the phonons in the film are therefore sensitive to this roughness. Moreover the surface defects responsible for the large scattering of phonons[21,22] can be regarded as two-level systems[23] (TLS). Their interaction with the film phonons leads to diffuse inelastic scattering of the phonons. We have therefore to consider a surface where the phonons undergo both specular and diffuse scattering with different probabilities.[24] The local specular reflection along with the surface roughness are both necessary to observe interference phenomena. The specular boundary scattering is characterized by a scattering rate $\tau_b^{-1}$,

$$\tau_b^{-1} = v/(\alpha w) , \tag{1}$$

where $v$ and $\alpha$ are, respectively, a mean phonon velocity and a parameter characterizing the specularity of the reflection.

The localized TLS's (Refs. 25 and 26) are coupled to the phonons through the deformation of the double-well potentials which is due to the elastic wave. The resonant absorption of a phonon $\hbar\omega$ by the TLS's (Ref. 27) in the vicinity of the surface is characterized by the phonon relaxation rate $\tau_{\text{ph-TLS}}^{-1}$:

$$\tau_{\text{ph-TLS}}^{-1}=\frac{3\pi}{\rho v^2}\gamma^2\bar{n}(E=\hbar\omega)\omega \ . \tag{2}$$

$E$ is the energy splitting of the two levels and $\gamma$ is the coupling energy between the phonons and the defects: we assume a single value[28] for longitudinal and transverse phonons $\gamma_L=\gamma_T=\gamma$; $\bar{n}(E=\hbar\omega)$ is an effective density of states per unit volume and per unit energy; $\rho$ is the mass density of the material and the factor 3 accounts for the three-phonon polarization with the mean velocity $v$ defined by

$$\frac{3}{v^2}=\frac{2}{v_T^2}+\frac{1}{v_L^2} \ , \tag{3}$$

$v_\alpha$ $(\alpha=T,L)$ being the sound velocity of the $\alpha$th polarization acoustic phonon averaged over the angles. Strictly speaking, the expression (2) for $\tau_{\text{ph-TLS}}^{-1}$ is valid for thermal phonons only $(\hbar\omega\sim kT)$. From the theory of tunneling states, the energy dependence of $\bar{n}(E)$ can be shown to be a logarithmic one,[26]

$$\bar{n}(E)\propto\ln(E/\Delta_{\min}) \ ,$$

where $\Delta_{\min}$ depends on the maximum acceptable barrier height $V_{\max}$ of the double potential well. Though the exact value of $V_{\max}$ is not known, the ratio $E/\Delta_{\min}$ can be evaluated for $V_{\max}=0.2$ eV;[25] it is shown to be very large. This leads to very slow variations of $\bar{n}(E)$ with $E$: therefore we assume an energy-independent density of states $\bar{n}(E)=n_0$. The resonant scattering of phonons by the TLS's is therefore characterized by an inelastic scattering rate proportional to the phonon angular frequency $\omega$,

$$\tau_{\text{ph-TLS}}^{-1}=B\omega \ . \tag{4}$$

## III. LOCALIZATION LENGTH AND INELASTIC PROCESSES

Following the work of Thouless[29] on electron localization, and a first extension to phonons in thin wires by Jäckle,[14] we will define a localization length for phonons in the thin films studied here. We consider phonon thermal diffusion along the length $L$ of the film; the phonon diffusion rate is determined in the usual way by the elastic scattering. As for electrons, the boundary conditions applied to the end of the film are relevant in the determination of the individual energy levels of the phonons. The time required for a phonon initially in a wave packet $(\omega)$ to diffuse from one end to the other end of the film can be written

$$t=(D_e/L^2)^{-1} \ , \tag{5}$$

where $D_e$ is the elastic diffusion constant.

The corresponding energy shift resulting from the uncertainty relation $\Delta\varepsilon=\hbar/t$ must be smaller than the spacing $\delta(\hbar\omega)$ between two adjacent energy levels determined by the boundary conditions in order that the phonon remains in the same energy level and wave packet. In two dimensions (2D), the spacing $\delta(\hbar\omega)$ is determined by the 2D density of states $n_2(\omega)$ per unit area and per unit frequency:

$$\delta(\hbar\omega)=\hbar/[n_2(\omega)Lw] \ .$$

From the relations

$$\Delta\varepsilon=\frac{\hbar}{t}=\frac{\hbar D_e}{L^2}<\frac{\hbar}{[n_2(\omega)Lw]}$$

a localization length $\xi\leq L$ can be defined by

$$\frac{D_e}{\xi^2}=\frac{1}{n_2(\omega)w\xi}$$

i.e.,

$$\xi=D_e n_2(\omega)w \ , \tag{6}$$

with

$$n_2(\omega)=\frac{1}{2\pi}\frac{3}{v^2}\omega \ , \tag{6'}$$

$v$ being the average phonon velocity defined in Eq. (3).

The diffusion constant $D_e$ is determined by the elastic scattering processes according to the relation

$$D_e=\tfrac{1}{2}vl_e \ , \tag{7}$$

where $l_e$ is the elastic mean free path; the factor $\frac{1}{2}$ is introduced here instead of the usual factor $\frac{1}{3}$ in three dimensions. The elastic processes we have to consider are first the boundary scattering with the rate $\tau_b^{-1}$ mentioned above [Eq. (1)] and then the Rayleigh scattering from the distribution of isotopic masses in a 2D system. The relaxation rate $\tau_{\text{iso}}^{-1}$ for the Rayleigh scattering can be deduced from the paper by Klemens[30] related to three-dimensional lattices. We obtain

$$\tau_{\text{iso}}^{-1}=S_0\tfrac{3}{8}\sum_i f_i\left[\frac{\Delta M_i}{M}\right]^2\frac{\omega^3}{v^2}=A\omega^3 \ , \tag{8}$$

where $A$ is frequency independent; $S_0$ is the surface per atom in a 2D lattice; $f_i$ is the concentration of isotope $i$ which mass differs from the mean atomic mass $M$ by the amount $\Delta M_i$. Equation (8) is established for an isotropic material.

The elastic scattering rate $\tau_e^{-1}$ and the corresponding mean free path $l_e=v\tau_e$ are obtained from Eqs. (1) and (8):

$$\tau_e^{-1}=\tau_b^{-1}+\tau_{\text{iso}}^{-1} \ .$$

We can define the localization time $\tau_\xi$ as the time required for a phonon to diffuse on the localization length with the elastic diffusion constant $D_e$:

$$\tau_\xi=\frac{\xi^2}{2D_e}=\frac{9}{16\pi^2}\frac{w^2}{v^2}\frac{\omega^2}{v/\alpha w+A\omega^3} \ . \tag{9}$$

This time is frequency dependent: at high frequency the Rayleigh scattering is dominant and the localization time decreases as $1/\omega$, whereas at low frequency, boundary scattering is dominant and $\tau_\xi$ increases as $\omega^2$.

The time $\tau_\xi$ must be smaller than the inelastic relaxation time $\tau_i$ characterizing the inelastic processes that make the phonon lose its phase coherence. We have already mentioned the TLS's scattering mechanism [Eqs.

(2) and (4)]. Another important inelastic mechanism is the spontaneous decay of phonons via three-phonon processes. It has been studied by Tamura[31] in bulk materials where decay rates proportional to $\omega^5$ are found for LA phonons. In contrast to bulk acoustic phonons, high-energy surface phonons exhibit very long lifetimes[32] in the range of seconds: this decay time corresponds to the interaction of surface phonons with bulk phonons and therefore is prohibited in free-standing films. We expect three-phonon processes in the free-standing film considered here to be still less efficient because of the reduced phase space. A quantitative estimate would require the consideration of Lamb acoustic waves in the film instead of the Rayleigh waves considered by Tamura.[33]

Therefore the single inelastic process than can prevent localization from occurring is the TLS's resonant scattering.

## IV. RESULTS AND DISCUSSION

We have calculated the localization time at $T=1$ K for a film width $w=1$ $\mu$m. The constants for Si (Ref. 31) have been used in the expression of $\tau_{\rm iso}^{-1}$ [Eqs. (8)]: $M_1=28$, $M_2=29$, and $M_3=30$, respectively, with the concentration $f_1=92.2\%$, $f_2=4.7\%$, and $f_3=3.09\%$. We obtain the mean velocity $v=6.09\times10^3$ ms$^{-1}$ from Eq. (3) with $v_L=9\times10^3$ ms$^{-1}$ and $v_T=5.4\times10^3$ ms$^{-1}$; $S_0=a^2/4$ where $a=5.43$ Å is the lattice constant of Si. With these constants, we get $A=0.15\times10^{-30}$ s$^2$. The inelastic scattering rate is estimated from Eq. (2). Only a mean value of the product $n_0\gamma^2$ can be measured experimentally.[34] Different values are found according to the material considered.[35,11] However, the order of magnitude of $\gamma$ can be evaluated by analogy with a distorted crystal.[26] Assuming $\gamma=1$ eV,[25,26,28] the effective density of states can be deduced from the value of $n_0\gamma^2$: with $n_0\gamma^2=3.9\times10^5$ J m$^{-3}$, $n_0=1.5\times10^{43}$ J$^{-1}$m$^{-3}$. With $\rho=2.33$ g cm$^{-3}$ (mass density of Si) and $v=6.09\times10^3$ ms$^{-1}$, we find $B=13\times\pi\times10^{-6}$ in Eq. (4).

In Fig. 1 are shown the $\omega$ dependences of $\tau_\xi^{-1}$ and of the inelastic scattering rate $\tau_{\text{ph-TLS}}^{-1}$ corresponding to the above values of $n_0$ and $\alpha$. The two curves cross one another at the frequency $\omega_0$. For phonons in the frequency range $\omega>\omega_0$, the phonon scattering by the TLS's is very efficient and prevents phonon localization from occurring ($\tau_i<\tau_\xi$) whereas localization is expected for phonons of frequency $\omega<\omega_0$. The value of $\omega_0$ depends on the parameter $\alpha$ that characterizes the partially specular feature of the reflection at the lateral surfaces and on the effective density of TLS's, $n_0$. For $\alpha=3$ and $n_0=1.5\times10^{43}$ J$^{-1}$m$^{-3}$ we find $\omega_0=6.5\times10^{11}$ s$^{-1}$: this frequency is larger than the value $\omega_{\rm th}=3.7\times10^{11}$ s$^{-1}$ for which the phonon energy density at 1 K is maximum. In these conditions, the localization of the modes $\omega<\omega_0$ is expected to reduce efficiently the thermal transport and to result in a small thermal conductivity. For larger values of $\alpha$, the localization time $\tau_\xi$ increases [Eq. (9)] and the TLS's are relatively more efficient; this results in a decrease of the frequency edge $\omega_0$ as is seen in Fig. 2(a): fewer frequency modes are localized. In Fig. 2(b), $\omega_0$ is

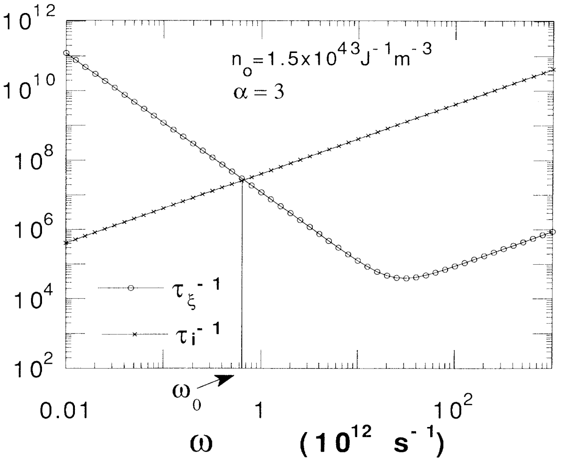

FIG. 1. Inverse localization time $\tau_\xi^{-1}$ and inelastic scattering rate $\tau_i^{-1}$ due to surface TLS's vs the phonon frequency $\omega$ for the reflection parameter $\alpha=3$ and the effective density of TLS's $n_0=1.5\times10^{43}$ J$^{-1}$m$^{-3}$; $\omega_0$ is the localization edge.

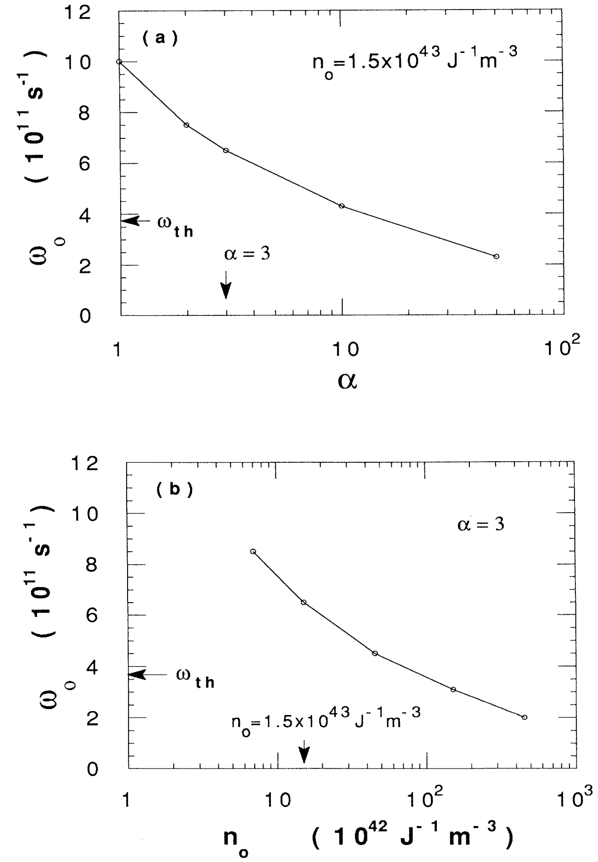

FIG. 2. Localization edge $\omega_0$; (a) as a function of the reflection parameter $\alpha$, for $n_0=1.5\times10^{43}$ J$^{-1}$m$^{-3}$; (b) as a function of the effective density of TLS's $n_0$, for $\alpha=3$. $\omega_{\rm th}$ is the thermal phonon angular frequency at 1 K.

seen to decrease with increasing $n_0$: larger effective density of the TLS's limits the localization to lower-frequency modes. In fact, to an increase of the specularity of the reflection and therefore to an increase of $\alpha$ must correspond a decrease of the number of defects at the source of the TLS's. An important feature of $\omega_0$ is that its order of magnitude lies in the range of the phonon thermal frequencies for real surface conditions, even with a value of $n_0$ smaller than the value considered above (Fig. 1).

In the material considered here, the value of $\omega_0$ is not immediately determined by the isotope scattering, since the crossover of the curves $\tau_g^{-1}(\omega)$ and $\tau_i^{-1}(\omega)$ is located in the frequency range where surface elastic scattering is dominant. The isotope concentration could be changed a lot without having a drastic effect on the localization edge $\omega_0$.

## V. CONCLUSION

Examination of the scattering processes in a free-standing two-dimensional film reveals that phonon localization is expected to occur for phonon modes $\omega$ smaller than a localization edge $\omega_0$. This edge lies in the frequency range of the phonon thermal frequencies at 1 K. The results presented above show that $\omega_0$ is essentially determined by the conditions of the film surfaces, which are real surfaces exhibiting roughness. In the frequency range $\omega < \omega_0$, the phonon modes do not participate in phonon heat transport; smaller thermal conductivity is therefore expected.[36] Measurements of the thermal conductivity of such films and comparisons with the conductivity expected from ideal surface films should give information on the surface condition in the free-standing films.

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[36] Quantitative estimates of the thermal conductivity will be done elsewhere.