Surface Brillouin scattering in polycrystalline gold

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Surface Brillouin spectra of supported gold films (500 and 930 Å) and of bulk gold have been measured with a tandem multipass Fabry-Perot interferometer. By fitting the calculated cross section to the experimental data the elastic and the photoelastic constants of gold have been derived. The surface spectrum of bulk gold shows, in addition to the Rayleigh wave peak, a broad resonance which is interpreted in terms of a leaky surface pseudomode of longitudinal character.

Opacity makes possible the use of Brillouin scattering to measure long-wavelength surface acoustic phonons in non-normal-incidence backscattering geometry.[1] In metals where the penetration depth of the light is very small, typically at least one order of magnitude smaller than the wavelength of the excitations, the light couples to acoustic vibrations exclusively through the ripple effect,[2] i.e., the dynamical corrugation of the surface induced by the thermal noise. In this case there is no coupling to bulk excitations through the usual elasto-optic mechanism. On the other hand, spectra taken of opaque semiconductors, like Si and GaAs, show at the same time surface and bulk features,[1] and the two above-mentioned scattering mechanisms coexist.

Gold shows an interesting intermediate behavior. In fact, its complex index of refraction $n+ik$ turns out to be equal to[3] $0.65+i1.90$, for light of wavelength $\lambda=5145$ Å. Although this peculiar value does not allow the bulk modes to be detected, it gives to the total scattering intensity from surface acoustic modes a significant elasto-optic contribution. These statements deserve a somewhat more detailed discussion. As far as the bulk modes are concerned it is well known[1] that the full width at half maximum of the bulk Brillouin peaks (neglecting phonon reflection at the boundary) is given by $\Delta\Omega=(2k/n)\Omega$ where Ω is the phonon frequency. For gold $\Delta\Omega/\Omega\simeq 6$ so that the bulk peaks are too broad to be detected in practice. The second statement, concerning the relative importance of the ripple and elasto-optic scattering mechanisms in surface Brillouin scattering, is not so obvious and the aim of this paper is indeed to elucidate this problem.

The Brillouin scattering cross section from surface waves in opaque materials is, in general, given by complicated expressions,[2,4,5] where the ripple and the elasto-optic contributions coherently mix together. Let us consider a semi-infinite medium in the half space $z<0$. The phonon wave vector is conveniently split into a part $\mathbf{Q}$ parallel to the surface and a normal part q. In the following we assume that $\mathbf{Q}$ is directed along the x axis. The phonon polarization vector $\mathbf{w}$ for a surface mode of frequency Ω and wave vector $\mathbf{Q}$ can be expanded in partial waves[6] as

$$\mathbf{w}(z)=\sum_{\lambda} a_{\lambda}\mathbf{v}_{\lambda}e^{iq_{\lambda}z} , \tag{1}$$

where λ labels the complex normal wave vectors at a given frequency Ω and surface wave vector $\mathbf{Q}$, $\mathbf{v}_{\lambda}$ is the polarization vector of the modes of the infinite medium, and a_{λ} are coefficients determined by the mechanical boundary conditions.[6] For a surface at $z=0$ the TM (transverse magnetic) $\rightarrow$ TM backscattering differential cross section has the form[5]

$$\frac{d^2\sigma}{d\Omega_a d\omega}\propto\sum_j\left|\sum_{\lambda} a_{\lambda}[A_{\lambda,x}v_{\lambda,x}+A_{\lambda,z}v_{\lambda,z}]+Bw_z(0)\right|^2 , \tag{2}$$

where Ω_a is the solid angle, ω is the frequency of the light, and j labels the independent solutions of the equation of motion of the semi-infinite crystal. The ripple contribution is given by the term $Bw_z(0)$, which is proportional to the normal component of the surface displacement field $w_z(0)$ times a constant B depending on the scattering angle θ and the dielectric constant ϵ of the medium. The elasto-optic term is more involved and is given by a complicated combination of longitudinal and transverse bulk displacements ($v_{\lambda,x}$ and $v_{\lambda,z}$, respectively) where the weighting factors $A_{\lambda,x}$ and $A_{\lambda,z}$ are linear combinations of the elasto-optic constants k_{ij} and are determined by the electromagnetic boundary contitions. In the more general case it is not immediately apparent from (2) what is the relative importance of the two scattering mechanisms, nor is it possible to unequivocally associate the displacement components with the two scattering mechanisms.

A great simplification occurs when $|\epsilon|$ is large. In this case it can be shown[5] that for an isotropic medium and TM $\rightarrow$ TM scattering

$$\frac{d^2\sigma}{d\Omega_a d\omega}\propto\sum_j|Aw_x(0)+Bw_z(0)|^2 , \tag{3}$$

where $A=\epsilon^{-3/2}(k_{11}-k_{12}c_{12}/c_{11})\sin\theta$ and $B=1+\sin^2\theta$.

This approximation was successful in explaining the surface spectrum of GaAs.[5,7] Equation (3) clearly associates the elasto-optic scattering with the longitudinal component w_x of the surface waves and the ripple scattering with the shear vertical component w_z. It is interesting to compare, in the case of gold, the different results given by the full theory (2) and by the approximation (3).

The ingredients for the calculation of the cross section are essentially the elastic moduli c_{ij} and the elasto-optic constants k_{ij}. These quantities are actually unknown for the gold films under consideration and have been directly determined from the Brillouin scattering experimental data.

Spectra have been measured for two thin films of polycrystalline evaporated gold with nominal thickness h, as determined by profilometer measurements, of about 500 and 1000 Å. The substrate was crystalline (001) silicon with **Q** aligned along [100]. A thick ($h \gg 1/Q$) free-standing film was also measured to compare with the calculated results for semi-infinite gold. This thick film was produced by evaporating 1000 Å of gold onto a glass substrate and subsequently electroplating more gold until the film separated naturally from the glass. The surface previously in contact with the glass was used for the measurement. A best-fit procedure to the measured spectra was used to determine both the elastic moduli and the elasto-optic constants. The results are summarized in Table I. Note that the fitting procedure is very sensitive to the film thickness. The values of 500 and 930 Å, respectively, obtained by fitting are considered more reliable than the profilometer measurements.

The experimental spectra were obtained with the tandem Fabry-Perot interferometer previously described.[1] An argon-ion laser ($\lambda = 5145$ Å) was used, with a power of 190 nW for the thin films and of 50 mW for the thick free-standing sample. The interferometer was used in a 3+3 multipass configuration with a scattering angle $\theta = 60°$, measured with respect to the surface normal.

The shear elastic modulus c_{44} of gold has been determined by fitting the frequency shift of the Brillouin peaks of the surface wave [Rayleigh wave (RW)] and of the guided waves in the supported films [Sezawa wave (SW) and Lamb waves of order n (LWn)]. The density ρ and the elastic modulus c_{11} of gold, given in Table I, have been taken from Ref. 3, because they are not accurately determined by the fit procedure. In fact, the dispersion curves of the guided wave modes, as a function of Qh, depend mainly on the transverse velocity of the film v_T. The elastic and photoelastic constants of the crystalline substrate, i.e., silicon, have been taken from the literature.[3,5] Our transverse velocity of gold $v_T = 1125$ m/s agrees within 5% with the value recently found by Brillouin scattering measurements on gold films deposited on glass.[8] The quality of the fitting is also comparable with that obtained in the latter reference: In both cases the overall agreement between calculated and measured values is within ±5%. The discrepancy is geater (7%) for the SW of the 500-Å sample at $Qh \simeq 1$. Notice that the same kind of discrepancy has been also found by Hillebrands *et al.*[8] for the SW of a 400-Å sample with nearly the same value of Qh.

Having determined the elastic properties of the polycrystalline gold used in our samples we are in a position to evaluate the absolute values of the elasto-optic constants of gold by fitting the shape of the experimental spectra. This is possible because, of the two independent mechanisms that contribute to the cross section, one of them (ripple) does not depend on the coupling constants k_{ij}. Therefore the constants k_{ij} can be found by fitting the amplitudes of the guided wave peaks (SW and LW) relative to the RW peak amplitude. These quantities are shown in Table I. The best fit values,

$$k_{11} = 42 - i4,\ k_{12} = 53 - i15,\ k_{44} = -5.5 + i5.5\ , \qquad (4)$$

are close (±5% in modulus) to those previously found with the same procedure for a set of different samples[9] obtained by evaporating gold on silicon. The fitting procedure has been carried out by using the full theory previously derived for Brillouin scattering from a support film.[10] The scattering from the substrate was not negligible for the 500-Å sample, where the penetration depth of the light in gold is comparable to the film thickness. The scattering from the 930-Å film took place in the film only.

The elasto-optic constants (4) have been used to com-

TABLE I. Experimental and calculated propagation velocities and Brillouin intensities for polycrystalline gold. The intensities are relative to the Rayleigh wave (RW). The elastic data of gold are also shown.

Sample		Velocity (m/s) RW	SW	LW1	LW2	Relative Brillouin intensity I_{SW}/I_{RW}	I_{LW1}/I_{RW}	I_{LW2}/I_{RW}
500-Å film	Measured data	2010	2530	4750		0.19	0.05	
	Fit data[a]	2018	2356	4678		0.258	0.04	
930-Å film	Measured data	1440	2180	3045	4250	0.21	0.11	0.007
	Fit data[a]	1481	2124	2981	4119	0.200	0.104	0.005
Bulk	Measured data	1065						
	Fit data[a]	1066						

[a]Elastic data of gold: $c_{11} = 2.07$ and $c_{44} = 0.25$ (10^{12} dyn/cm^2), $\rho = 19.75$ (g/cm^3). The intensities are calculated with the full theory (2) and the elasto-optic constants (4).

pute the surface Brillouin spectrum of semi-infinite gold. The comparison between the calculated and experimental results is shown in Fig. 1. The surface spectrum is characterized by a peak due to the RW and by a continuum due to the bulk modes reflected at the boundary. It must be emphasized that this continuum, although originated by bulk modes, has an explicit surface character and has nothing to do with the true bulk resonances, not observable in gold as mentioned before.

In the calculated spectrum of Fig. 1 the RW peak (originally a Dirac δ function) has been given a finite width at half maximum, to compare with the experimental results. The agreement between the theory and the experiment is very good. The continuum spectrum is of particular interest. In gold it is markedly different from that of strongly absorbing metals, as aluminium and nickel,[1] which show a plateau between Ω_T and Ω_L due to the ripple coupling with shear vertical modes. It is also different from the continuum spectrum of GaAs[1] which on the contrary shows a well-defined peak located at Ω_L. One can see in Fig. 1 that the surface spectrum of gold is characterized by a large bump located in between the two thresholds. In order to investigate the nature of this structure we compare in Fig. 2 the total calculated cross section with the cross section computed by retaining either the ripple or the elasto-optic contribution in expression (2). Although the elasto-optic term is almost everywhere smaller than the ripple one, it is by no means negligible and is essential in order to understand the behavior of the cross section between Ω_T and Ω_L. Notice that the two mechanisms coherently interfere and give rise to cancellation at low frequency, whereas they add for $\Omega \geq 7$ GHz. Obviously this varying character of the interference is due to the complex nature of the scattering amplitudes appearing inside the square modulus of Eq. (2). The interference process turns out to accurately reproduce both the position of the bump in the continuum and its relative intensity with respect to the RW. From the analysis of Fig. 2 it can be concluded that the bump is of prevailing elasto-optic nature and that the interference enhances its intensity and shifts its maximum to a lower frequency, 0.8 GHz below.

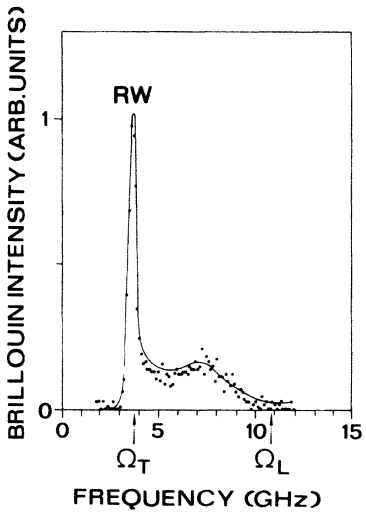

FIG. 1. Measured (points) and calculated (solid line) surface Brillouin spectra of polycrystalline gold. RW: Rayleigh wave peak. Ω_T and Ω_L are the transverse and longitudinal thresholds of bulk modes, respectively. The relative intensity of the two spectra has been fixed by fitting the amplitude of the RW.

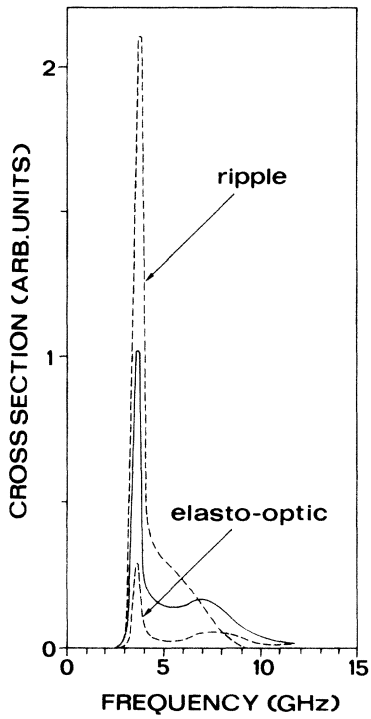

FIG. 2. Total calculated cross section (solid line) compared with the ripple and elasto-optic cross section (dashed lines). The full theory (2) has been used. The curves are drawn in the same scale.

The cross-section calculation of gold has been also carried out with the approximate formula (3). The results, shown in Fig. 3, are qualitatively similar to those of Fig. 2, but the detailed behavior is different. In the total cross section the ratio between the RW and the continuum is too

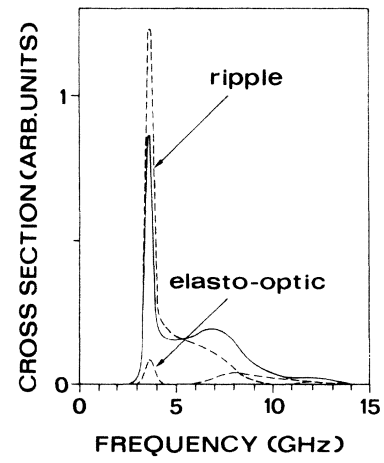

FIG. 3. As in Fig. 2, but the approximate expression (3) has been used. Figures 2 and 3 have been drawn to the same scale.

small and does not agree with the experimental data. Figure 3 is a useful guide to interpret the physical meaning of the bump in the continuum. In fact, it shows that this structure owes its existence to the behavior of the longitudinal displacements $w_x(0)$ between Ω_T and Ω_L and that the elasto-opitc coupling is mainly responsible for the appearing of the bump. The broad peak in the displacements is due to a surface pseudomode of longitudinal character, or leaky mode.[7,11]

It must now be explained why in polycrystalline gold the leaky mode is located at a frequency $\Omega < \Omega_L$ and why this resonance is broader than in GaAs.[5,7] The explanation may be found in the high value of the Poisson ratio of gold $\sigma \simeq 0.43$. In a previous paper[12] the shape of the longitudinal displacement component $w_x(0)$ has been studied in detail as a function of σ. When $\sigma \geq \frac{1}{3}$ it has been found that $w_x(0)$ shows a broad resonance in the continuum spectrum located at about $2\Omega_T$, at a frequency smaller than Ω_L. Although we have shown that the elasto-optic scattering amplitude is not simply proportional to $w_x(0)$, the superposition of partial waves entering Eq. (2) produces a resonance in the elasto-optic scattering amplitude similar to that exhibited by $w_x(0)$, as can be seen by comparing Figs. 2 and 3. However, in order to get the correct cross section, the approximate expression (3) is not sufficiently accurate, because of the fine interference between the two terms which requires a detailed knowledge of both the amplitude and phase of the ripple and elasto-optic contributions.

In conclusion, we have shown that the Brillouin spectrum of gold exhibits an interesting feature betwen Ω_T and Ω_L, which is the fingerprint of the leaky longitudinal mode[11,12] of the surface acoustic spectrum of solids.

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