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Theory of Brillouin scattering on a surface grating: Role of surface polaritons

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In a recent paper [W. M. Robertson *et al.*, Phys. Rev. B **41**, 4986 (1990)] an empirical and unexplained selection rule governing light scattering by surface acoustic waves on corrugated metal surfaces has been found experimentally, whenever a surface polariton acts as in intermediate state given rise to a *Rayleigh wave replica* in the spectra. The *replica* occurs only when the scattered light is s polarized, independently of the incident polarization. In this paper the Brillouin cross section for a corrugated surface is evaluated theoretically within the extinction-theorem formalism and the above-mentioned selection rule is shown to be valid in the limit of large dielectric constant.

Due to technical progress in high-resolution and high-contrast spectrometers, Brillouin scattering has become an important tool to investigate the dynamical properties of nontransparent solids.[1,2] When the light penetration depth is comparable with the phonon wavelength, the incident light is scattered inelastically by the fluctuating dielectric inhomogeneities generated by thermal waves (elasto-optic coupling)[3,4] and gives information on the phonon modes in the region underneath the surface. For an opaque material such as a metal, where the light penetration depth becomes very small, the scattering occurs only in the vicinity of the surface, thus making Brillouin scattering particularly suited to study long-wavelength surface phonons.[5,6] In the latter case the coupling occurs mainly via the ripple mechanism[6-13] which acts as a dynamical grating still causing a shift in frequency (Doppler effect)

$$\omega_f=\omega_i\pm\Omega \tag{1}$$

as well as in momentum parallel to the surface

$$\mathbf{K}_f=\mathbf{K}_i\pm\mathbf{Q}\,. \tag{2}$$

The signs + and − refer to absorption or emission of a phonon of energy Ω and parallel momentum $\mathbf{Q}$ (anti-Stokes and Stokes processes), and $\mathbf{K}_{f,i}$ denote the scattered and incident parallel momentum of the photon,

$$K_f=(\omega_f/c)\sin\theta_f\,, \tag{3}$$

$$K_i=(\omega_i/c)\sin\theta_i\,, \tag{3'}$$

where θ_f and θ_i are the azimuthal angles of the scattered and incident light, respectively. As in Ref. 11, here and in the following sections we use capital letters for vectors in the (x,y) surface plane, thus $\mathbf{r}\equiv(\mathbf{R},z)$ and $\mathbf{R}\equiv(x,y)$, accordingly.

Recently in the experiments by Robertson *et al.*[14,15] surface Brillouin spectra have been measured on a silver grating rather than on a flat surface. The introduction of a grating has two consequences. First, it modifies the phonon spectrum since it mixes phonon modes with the same frequency Ω but different momenta $\mathbf{Q}$, $\mathbf{Q}'$ with $\mathbf{Q}'=\mathbf{Q}+\mathbf{G}$, where $\mathbf{G}$ is a two-dimensional (2D) grating momentum. Second, it generates diffraction coupling the light channels $\mathbf{K}$, $\mathbf{K}'$, with $\mathbf{K}'=\mathbf{K}+\mathbf{G}$. The first fact causes an Ω *gap* in the Rayleigh wave at the Brillouin-zone edge;[16] the second makes possible the light coupling with the surface polariton (SP) which can occur at particular angles θ_i, θ_f. This second effect has been observed by Robertson *et al.*[15] and gives rise to a *Rayleigh replica* in their spectral intensity. The strange thing, however, is that this second peak appearing at a higher frequency is present only for s-polarized scattered light while it is absent for p polarization.[15,17] Moreover, the result turns out to be independent of the incident polarization. From these findings the authors of Ref. 15 suggest an empirical selection rule as in their Table I which can hardly be accepted to hold in general. In fact, for in-plane scattering and perfect antireflection

$$\mathbf{K}_f=-\mathbf{K}_i \tag{4}$$

the time-reversal symmetry imposes for the cross section

$$\sigma_{p\leftarrow s}=\sigma_{s\leftarrow p}\,. \tag{5}$$

The latter equation shows the equality of the mixed polarization intensities, thus the breakdown of the empirical selection rule for this particular channel.

A quantitative comparison with the experiment can be done extending the plane surface theory of Ref. 11 to a grating. This is accomplished generalizing the *extinction theorem* formalism[18-22] to a dynamical grating

$$z=\zeta(\mathbf{R}-\mathbf{u}_\parallel)+u_z\,, \tag{6}$$

where the shear-vertical phonon displacement u_z gives rise to the ripple contribution. The elasto-optic contribution can be included on the same footing. In Eq. (6)

$$z=\zeta(\mathbf{R}) \tag{7}$$

defines the (static) 2D grating surface and $\mathbf{u}\equiv(\mathbf{u}_\parallel,u_z)$ is the elastic displacement evaluated at the surface grating.

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Linearizing these equations in the elastic displacements and omitting the details which will be given elsewhere[23] we get the Brillouin cross section

$$\left.\frac{d^2\sigma}{d\omega_f d\Omega_f}\right|_{\alpha\leftarrow\beta} = \frac{1}{(2\pi)^2}\left(\frac{\omega_f}{c}\right)^2 \frac{\cos^2\theta_f}{\cos\theta_i}\,\frac{\hbar\tilde{N}(|\omega_f-\omega_i|)}{2\rho|\omega_f-\omega_i|}\sum_{n,\mathbf{Q},\mathbf{G}}\delta(\Omega_n-|\omega_f-\omega_i|)\,\delta_{\mathbf{K}_f-\mathbf{K}_i,\mathbf{Q}+\mathbf{G}}\,|(A_{K_f,\omega_f}[\mathbf{w}^n_{\mathbf{Q}}])_{\alpha,\beta}|^2 . \quad (8)$$

The 2×2 matrix A contains the amplitude of the electric field and $\alpha,\beta=1,2$ denote the p and s polarization of the scattered and incident fields. Here we use the nomenclature of Ref. 11 and A is a linear functional of the normal mode $\mathbf{w}^n_{\mathbf{Q}}\equiv\mathbf{w}^n(z,\mathbf{R};\mathbf{Q})$, which is a Bloch solution of the 2D grating given by Eq. (7). We represent it in the extended zone scheme to make a closer connection with the plane surface result.[24] With this choice n still includes the polarization index of phonon modes on the degenerate energy level Ω_n. Contrary to the plane surface result, Eq. (8) shows that all normal modes with momenta

$$\mathbf{Q}+\mathbf{G}=\mathbf{Q}_d\equiv\mathbf{K}_f-\mathbf{K}_i \quad (9)$$

are contributing in the $\mathbf{K}_f,\omega_f\leftarrow\mathbf{K}_i,\omega_i$ transition, $\mathbf{G}$ being borrowed by the static grating of Eq. (7).

The expression for $A_{\mathbf{K}_f,\omega_f}[\mathbf{w}^n_{\mathbf{Q}}]$ is easily found once we solve the equation for

$$A(\mathbf{K}_f,\omega_f,\mathbf{K}_i,\omega_i)=\sum_{\mathbf{Q}}A_{\mathbf{K}_f,\omega_f}[\mathbf{w}^n_{\mathbf{Q}}]\,|_{\Omega_n=|\omega_f-\omega_i|} \quad (10)$$

since it is linear in the displacements $\mathbf{w}^n_{\mathbf{Q}}$. Neglecting the *elasto-optic* contribution and using the T-matrix formalism as in Brown *et al.*,[19,21] one has the following, unless elastic reflection is present,

$$A(\mathbf{K},\omega,\mathbf{K}_i,\omega_i)=-2ip(\mathbf{K}_i,\omega_i)G^0(\mathbf{K},\omega)\times\tilde{A}(\mathbf{K},\omega,\mathbf{K}_i,\omega_i)G^0(\mathbf{K}_i,\omega_i) \quad (11)$$

and

$$\tilde{A}=V^u+V^\zeta G V^u+V^u G V^\zeta , \quad (12)$$

with

$$G=G^0+G^0V^\zeta G . \quad (13)$$

In the above equations we have used a compact notation and the equations have been linearized in V^u vertex which contains the *ripple* contribution. The static part of the corrugation (grating) is contained in V^ζ defined as

$$V^\zeta(\mathbf{K},\omega,\mathbf{K}',\omega')=\delta_{\omega,\omega'}V^\zeta_\omega(\mathbf{K},\mathbf{K}') . \quad (14)$$

Lowest-order solutions for V^ζ,V^u are

$$V^\zeta_\omega(\mathbf{K},\mathbf{K}')=\zeta_{\mathbf{K}-\mathbf{K}'}U(\mathbf{K},\mathbf{K}') , \quad (15)$$

$$V^u(\mathbf{K},\omega,\mathbf{K}',\omega')=w^n_z(z=0,\mathbf{K}-\mathbf{K}')\,|_{\Omega_n=|\omega-\omega'|}U(\mathbf{K},\mathbf{K}') , \quad (15')$$

where

$$U(\mathbf{K},\mathbf{K}')=\frac{(\epsilon-1)}{\epsilon}\begin{pmatrix} KK'-\hat{\mathbf{K}}\cdot\hat{\mathbf{K}}'qq'/\epsilon & -(\omega/c)q\hat{\mathbf{K}}\times\hat{\mathbf{K}}'\cdot\hat{\mathbf{z}} \\ -(\omega/c)q'\hat{\mathbf{K}}\times\hat{\mathbf{K}}'\cdot\hat{\mathbf{z}} & \epsilon(\omega/c)^2\hat{\mathbf{K}}\cdot\hat{\mathbf{K}}' \end{pmatrix} , \quad (16)$$

with $\hat{\mathbf{K}}$, $\hat{\mathbf{K}}'$ being unit vectors. Equation (15) agrees with Eq. (1.7) of Ref. 21, while in Eq. (15′) $w^n_z(z;\mathbf{Q})$ is the normal mode for a flat surface.[24] The coupling of modes due to grating has been consistently neglected to lowest order. Still in (15′) we have used

$$|\omega-\omega'|=\Omega_n\ll\omega,\omega' \quad (17)$$

and neglected terms $\sim O(\Omega_n/\omega)$. The quantity G^0 in Eqs. (11) and (13) is the Green's function for the plane surface[21]

$$G^0_{\alpha\beta}(\mathbf{K},\omega)=\delta_{\alpha,\beta}G^0_\alpha(\mathbf{K},\omega) , \quad (18)$$

$$G^0_1(\mathbf{K},\omega)=i\epsilon/(q+\epsilon p),\quad G^0_2(\mathbf{K},\omega)=i/(q+p) , \quad (18')$$

where

$$p\equiv p(\mathbf{K},\omega)=[(\omega/c)^2-K^2]^{1/2} ,$$
$$q\equiv q(\mathbf{K},\omega)=[\epsilon(\omega)(\omega/c)^2-K^2]^{1/2} , \quad (19)$$

with $\mathrm{Im}q,p\geq0$. Equation (13) is thus the Green's-function equation for a grating. Rewriting it as

$$G=G^0+G^0V^\zeta G^0+G^0V^\zeta G^0V^\zeta G \quad (20)$$

the equation can be solved by neglecting the off-diagonal elements:

$$G_{\alpha,\beta}(\mathbf{K},\omega)=\delta_{\alpha,\beta}G_\alpha(\mathbf{K},\omega) , \quad (21)$$

$$G_\alpha^{-1}(\mathbf{K},\omega)=G^{0^{-1}}_\alpha(\mathbf{K},\omega)-\sum_{\mathbf{K}'}\sum_{\gamma=1,2}V^\zeta_{\alpha,\gamma}(\mathbf{K},\mathbf{K}')G^0_\gamma(\mathbf{K}',\omega)V^\zeta_{\gamma,\alpha}(\mathbf{K}',\mathbf{K}) , \quad (22)$$

and from

$$G_1^{-1}(\mathbf{K},\omega)=0 \quad (23)$$

one recovers back the SP dispersion relation on a grating in a small corrugation limit.[16]

Selecting the surface polariton contribution in Eq. (12) one still has

$$\tilde{A}_{\alpha\beta}\simeq V^u_{\alpha\beta}+V^\zeta_{\alpha1}G_1V^u_{1\beta}+V^u_{\alpha1}G_1V^\zeta_{1\beta} . \quad (24)$$

Equation (12) or Eq. (24) reduce for

$$G\rightarrow G^0 \quad (25)$$

to the second-order perturbation theory result. Contrary to this they both include the SP damping due to grating and which cannot be neglected if $\mathrm{Im}\epsilon\ll1$ as in the case of silver. The first term on the right-hand side (rhs) in Eq. (24) is the *direct* transition and in Eq. (8) it gives back the *ripple* contribution to the Brillouin cross section for a flat surface.[11] The other two terms are schematically represented in Figs. 1(a) and 1(b), respectively. We have supposed, as in the experiment by Robertson *et al.*,[15] that the scattering occurs in the sagittal plane parallel to the grating grooves of momentum $\mathbf{G}$. The phonon involved in the scattering has energy $\Omega_n = \omega_f - \omega_i$ and momenta $\mathbf{Q}_1$, $\mathbf{Q}_2$, while the resonance with the SP occurs if, as in Fig. 1(a),

$$|\mathbf{K}_f - \mathbf{G}| = K_{\mathrm{SP}} \tag{26}$$

or, as in Fig. 1(b)

$$|K_i + G| = K_{\mathrm{SP}} , \tag{27}$$

where K_{SP} is given by Eq. (23), thus approximately

$$K_{\mathrm{SP}} = \left(\frac{\omega}{c}\right) \mathrm{Re} \left(\frac{\epsilon}{\epsilon+1}\right)^{1/2} \tag{28}$$

the SP momentum for a flat surface.

In the experment by Robertson *et al.*[15] the final channel is taken in such a way that Eq. (26) is satisfied, while Eq. (27) does not hold in general, $\mathbf{K}_i$ being arbitrary. This means that in Eq. (24) the second term on the rhs is resonant and prevailing over the third, which is thus irrelevant. Of course the situation would be opposite if Eq. (27) held and Eq. (26) did not. In the latter case it would be the second term in Eq. (24) to be dropped. Using Eqs. (15), (15′), and (16) and selecting only resonant contributions, we can write compactly

$$\begin{aligned}\tilde{A}_{\alpha\beta}(\mathbf{K}_f,\omega_f,\mathbf{K}_i,\omega_i) \simeq\ & w_z^n(0;\mathbf{Q}_d) U_{\alpha\beta}(\mathbf{K}_f,\mathbf{K}_i) - \zeta_{\mathbf{G}} w_z^n(0;\mathbf{Q}_d - \mathbf{G}) \\ & \times G_{\mathrm{SP}}[M_{\alpha\beta}(K_f,K_i,G)\delta_{|\mathbf{K}_f-\mathbf{G}|,K_{\mathrm{SP}}} + M_{\beta\alpha}(K_i,K_f,-G)\delta_{|\mathbf{K}_i+\mathbf{G}|,K_{\mathrm{SP}}}] \\ & - \zeta_{-\mathbf{G}} w_z^n(0;\mathbf{Q}_d+\mathbf{G}) G_{\mathrm{SP}}[M_{\alpha\beta}(K_f,K_i,-G)\delta_{|\mathbf{K}_f+\mathbf{G}|,K_{\mathrm{SP}}} + M_{\beta\alpha}(K_i,K_f,G)\delta_{|\mathbf{K}_i-\mathbf{G}|,K_{\mathrm{SP}}}] ,\end{aligned} \tag{29}$$

where $\mathbf{Q}_d = \mathbf{K}_f - \mathbf{K}_i$ as in Eq. (9) and

$$M(K_f,K_i,G) = \frac{(\epsilon-1)^2}{\epsilon} \begin{pmatrix} K_f(\gamma_{\mathrm{SP}}-\gamma_f)/\epsilon \\ -iG\omega/c \end{pmatrix} \left(\frac{K_i\gamma_{\mathrm{SP}}+K_f\gamma_i}{\epsilon}, -iG\omega/c\right) . \tag{30}$$

Still G_{SP} is defined in Eq. (22) with $\mathbf{K} = \mathbf{K}_{\mathrm{SP}}$ and gives

$$G_{\mathrm{SP}}^{-1} = \frac{\gamma_{\mathrm{SP}}}{\epsilon} + \beta_{\mathrm{SP}} - \left(\frac{\epsilon-1}{\epsilon}\right)^2 \sum_{\mathbf{K}'} |\zeta_{\mathbf{K}-\mathbf{K}'}|^2 \frac{(\mathbf{K}_{\mathrm{SP}}\cdot\mathbf{K}' - \gamma_{\mathrm{SP}}\beta')(\mathbf{K}_{\mathrm{SP}}\cdot\mathbf{K}' - \gamma'\beta_{\mathrm{SP}})}{\gamma'/\epsilon + \beta'} , \tag{31}$$

with $\gamma = -iq$, $\beta = -ip$ where q, p are defined by Eq. (19). From Eqs. (8), (10), (11), and (29) one determines the Brillouin cross section. For a perfect antireflection, defined by Eq. (4), and mixed polarization scattering, the *direct* contribution—the first term in Eq. (29)—vanishes and one recovers Eq. (5), which is indeed a general property for this scattering channel. Vice versa if, as in the experiment,[15] only Eq. (26) is valid, we have, neglecting for a moment the *direct* contribution,

$$\left.\frac{\sigma_{p\leftarrow\beta}}{\sigma_{s\leftarrow\beta}}\right|_{\mathrm{replica}} = \left| \frac{G_1(\mathbf{K}_f,\omega_f)}{G_2(\mathbf{K}_f,\omega_f)} \frac{K_f(\gamma_{\mathrm{SP}}-\gamma_f)}{\epsilon(\omega/c)G} \right|^2 \tag{32}$$

independent of the incident polarization. The equation above gives the ratio of the peak intensities for the *Rayleigh replica.* The contribution of the *direct* term involves a phonon mode in the continuum and it is thus negligible.

For a laser beam with $\lambda = 5145$ Å as in the experi-

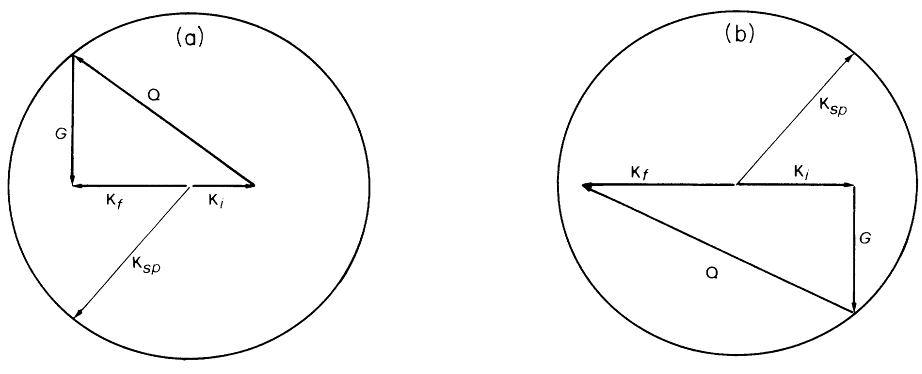

FIG. 1. Resonant SP conditions for an *in-plane* scattering geometry where the scattering plane is parallel to the grating grooves. (a) The resonance occurs via the phonon with a parallel momentum of $\mathbf{Q} = \mathbf{Q}_d - \mathbf{G}$ where $\mathbf{Q}_d = \mathbf{K}_f - \mathbf{K}_i$ as in Eq. (11). (b) The resonance occurs via the grating with a momentum of $\mathbf{G}$. Since $\mathbf{Q}_d$ and $\mathbf{G}$ are orthogonal vectors, one gets similar resonant processes, not shown in the figures, with $\mathbf{G} \rightarrow -\mathbf{G}$ in the lower half plane of (a) and in the upper half plane of (b).

ment,[15] we take $\epsilon_1 = -10.5$, $\epsilon_2 = 0.33$ as the real and imaginary parts of the dielectric constant for the Ag grating.[25] Using

$$G^2 = K_{SP}^2 = K_f^2 \tag{33}$$

and

$$\gamma_{SP} - \gamma_f \simeq \frac{1}{2}\frac{\omega}{c}\frac{\cos^2\theta_f}{(|\epsilon_1|)^{1/2}} \tag{34}$$

valid for $|\epsilon_1| \gg 1$, we have from Eq. (32) the final result

$$\left.\frac{\sigma_{p\leftarrow\beta}}{\sigma_{s\leftarrow\beta}}\right|_{\text{replica}} = \frac{|\epsilon_1|-1}{|\epsilon_1|}\frac{\sin^2\theta_f\cos^4\theta_f}{4(\sin^2\theta_f + |\epsilon_1|\cos^2\theta_f)^2} \tag{35}$$

$$\simeq \frac{\sin^2\theta_f}{4|\epsilon_1|^2} \; . \tag{35'}$$

The latter equation is in agreement with the experimental data.[15] The *effective* selection rule arises in this case from the large value of the dielectric constant and this result can be retained valid for many metals in the optical range.

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