# Light scattering by surface acoustic waves on corrugated metal surfaces

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We report the results of a Brillouin-scattering study of corrugated Ag surfaces. The corrugation plays a dramatic role in the wave-vector-selection rules governing coupling to surface phonons, and this effect is substantially different when the effective wave vector of the surface corrugation is collinear or perpendicular to the scattering plane. In processes that involve the grating wave vector, we show that the coupling mechanism between light and phonons is governed by surface plasmons which introduce a new scattering interaction with unusual polarization features in the Brillouin-scattering process.

#### I. INTRODUCTION

The scattering of light by surface acoustic waves on opaque materials is a well-known phenomenon.<sup>1-4</sup> It has been found that the sinusoidal ripple produced at the surface by a propagating surface acoustic wave acts as a "moving grating" which can diffract incident light. The fact that the "grating" created by the propagating acoustic wave is moving with the surface-acoustic-wave velocity results in a Doppler shift in the diffracted light. This frequency shift can be measured using Brillouinscattering techniques.<sup>2</sup> Scattering of light by the ripple mechanism has been shown to be the dominant lightscattering mechanism by acoustic phonons in opaque materials, and theoretical and experimental details of the mechanism have been extensively described in the literature.<sup>5-7</sup> In scattering experiments from surface phonons the wave vector of the phonon  $(\mathbf{q})$  involved in the scattering process is determined by the conservation of the components of the wave vectors of the incident  $(\mathbf{k}_i)$  and scattered  $(\mathbf{k}_{s})$  light parallel to the surface. If the incident and scattered light subtend angles  $\theta_i$  and  $\theta_s$  with respect to the surface normal ( $\theta_i$  and  $\theta_s$  assumed to lie in the same plane), then the surface phonon responsible for scattering has a wave vector

$$q_0 = k_i \sin \theta_i + k_s \sin \theta_s \ . \tag{1}$$

Because surface Brillouin scattering is essentially a process of diffraction of the incident light by the corrugation created by the surface phonon, the polarization of the scattered light is the same as that of the incident light.

In extending Brillouin-scattering experiments to corrugated surfaces a number of effects are to be expected. The intrinsic phonon characteristics (e.g., dispersion relations, amplitude, etc.) may be altered by the presence of a surface corrugation. Calculations of phonon propagation on corrugated surfaces have been reported,<sup>8,9</sup> and these calculations indicate that, provided the amplitude of the corrugation is small ( < 10% of the grating spacing), the dispersion relation of the Rayleigh surface acoustic wave is not found to be greatly affected. Furthermore, when changes are observed, they are most pronounced for phonons with wavelengths close to multiples of the corrugation period. In a previous investigation<sup>10</sup> of phonons propagating along the direction of corrugation (i.e., perpendicular to the grooves) our results were found to be consistent with the behavior predicted in Refs. 8 and 9.

Another feature which could be expected on a corrugated surface is the breakdown of Eq. (1). If the corrugation is described by an effective wave vector  $\mathbf{k}_g = 2\pi/d$ (where d is the grating spacing) then it is reasonable to expect, from a consideration of parallel wave-vector conservation, that it is possible to couple to all phonons which satisfy

$$q = k_i \sin \theta_i + k_s \sin \theta_s + nk_g = q_0 + nk_g \tag{2}$$

with n an integer. In Ref. 10 we investigated the case when **q** is parallel to  $\mathbf{k}_g$  and, for an arbitrary angle of incidence, found no evidence of a modification of Eq. (1). However, if the angle of incidence of the light was chosen to be such that surface plasmons were generated, then the wave-vector-conservation condition given by Eq. (2) was clearly observable.

Here we report results of experiments performed with  $k_g$  not colinear with q. The extension of Eq. (2) to the general case is derived and we find that it is necessary to use this generalized expression for a wide range of incident angles. Further, the polarization features of the scattered light are surprising but can be accounted for by considering surface plasmons as real intermediate states in the scattering process.

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#### **II. EXPERIMENT**

Brillouin spectra were recorded on a (5+4)-pass Fabry-Perot interferometer<sup>2</sup> and laser powers in the range 20-200 mW of 514.5-nm radiation were used. In our experiments the surface normal **n** was horizontal, and we refer to the polarization of the incident and scattered light as *H* (the electric field in the scattering plane) and *V* (vertical). This choice of notation is made to avoid the ambiguity of the more usual "s" and "p" notation which could be interpreted either with respect to the scattering plane or the grating plane (viz., a plane containing  $\mathbf{k}_g$  and **n**).

The gratings used in these experiments have been described in detail in Ref. 10; they consist of 300 nm of Ag sputtered onto sinusoidally corrugated photoresist which were holographically produced. The gratings utilized had corrugation amplitudes of 10.6, 23.8, 30.4, and 33 nm, and the wavelength of the corrugation was 720 nm on all samples. The experiments were carried out in a configuration in which the surface normal and the incident and scattered directions are coplanar. The angle between the surface normal and  $\mathbf{k}_i$  and  $\mathbf{k}_s$  are  $\theta_i$  and  $\theta_s$ , respectively. The angle between the grating wave vector,  $\mathbf{k}_g$ , and the scattering plane is labeled  $\beta$  such that  $\beta=0^\circ$ corresponds to the previously reported case of **q** parallel to  $\mathbf{k}_g$ .

### **III. RESULTS**

In this section we first contrast the Brillouin-scattering results for  $\beta = 0^{\circ}$  to those for  $\beta = 90^{\circ}$  and show that the frequency shifts of the observed peaks can be accounted for by a generalization of the wave-vector conservation relation of Eq. (2). Figure 1(a) shows a Brillouin spectrum taken with  $\beta = 0^{\circ}$ , i.e., with the grating wave vector and the scattering phonon collinear as shown in the inset to Fig. 1(a), and with incident light H polarized. This corresponds to the scattering configuration described in Ref. 10 and as detailed there we found, for light incident at angles away from the surface-plasmon resonant angle, no difference in the Rayleigh surface acoustic velocity between the corrugated and uncorrugated surfaces. In the spectrum shown the large truncated peak around zerofrequency shift is due to the elastically scattered laser light while the two sharp peaks at higher frequency are due to light scattered by the Rayleigh surface acoustic wave and by the first Sezawa mode, a guided acoustic wave of the 300-nm Ag film.

The spectrum in Fig. 1(b) for the case  $\beta = 90^{\circ}$  is markedly different from that shown in Fig. 1(a). The incident and scattered angles are the same as for Fig. 1(a) and the Rayleigh and Sezawa peaks are still present; however, there are two new peaks: one at a frequency between that of the Rayleigh and Sezawa modes and the other at a frequency shift larger than the Sezawa mode. These modes appear to be "split off" from the regular Rayleigh and Sezawa peaks. To see if these split-off modes have the character of true surface waves their frequency shift is plotted as a function of the scattering phonon wave vector  $q_0$  defined by Eq. (1). The resulting dispersion curve is shown in Fig. 2. The true (open symbols) Rayleigh (circles) and Sezawa (squares) modes have the expected behavior with q, i.e., the same dispersion as for the case  $\beta = 0^{\circ}$ ; the split-off modes (solid symbols), however, tend to merge with the regular Rayleigh and Sezawa modes at large phonon wave vectors. This behavior can be understood by considering a generalization of Eq. (2) for the case in which  $\mathbf{k}_g$  is not in the plane of  $\mathbf{k}_i$ and  $\mathbf{k}_s$ . Figure 3 shows a vector diagram, in  $\mathbf{k}$  space parallel to the surface, for the grating-relaxed scattering. The scattering wave vectors  $q^{\pm}$  are given by

$$q^{\pm} = [(k_i \sin\theta_i + k_s \sin\theta_s \pm k_s \cos\beta)^2 + k_g^2 \sin^2\beta]^{1/2}$$
$$= [(q_0 \pm k_g \cos\beta)^2 + k_g^2 \sin^2\beta]^{1/2}$$
(3)



FIG. 1. Brillouin spectra taken from Ag-covered grating with incident polarization H, no analyzer. (a)  $\beta = 0^{\circ}$  showing the conventional Rayleigh and Sezawa Brillouin peaks. (b)  $\beta = 90^{\circ}$ showing the conventional Rayleigh and Sezawa peaks and the split-off modes. The insets to each figure show the relative orientation of the grating grooves with respect to the phonon responsible for scattering. The phonon wave vector **q** is determined by the incident and scattered light directions as described by Eq. (1).

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2.6

relaxed condition was observed *only* if the incident light was directly coupled to surface plasmons. Another surprising observation is the polarization characteristics of the scattered radiation. The spectra in Fig. 1 were taken using the conventional surface Brillouin-scattering arrangement which uses incident light with horizontal (H)polarization and no polarization analyzer in the scattered



FIG. 2. Measured dispersion curve for the conventional (open symbols) Rayleigh (circles) and Sezawa (squares) modes and for the split-off modes (solid symbols).

2.4

PHONON WAVE VECTOR (  $10^{-6} \text{ m}^{-1}$  )

2.2

which reduces to Eq. (2) for  $\beta = 0^{\circ}$  and which for  $\beta = 90^{\circ}$  can be simplified to

$$q = (q_0^2 + k_g^2)^{1/2} . (4)$$

Clearly the involvement of the grating wave vector in the scattering process leads qualitatively to the observed q dependence because with larger wave-vector transfer, q, the effect of  $k_g$  is diminished and the split-off modes tend to merge with the true Rayleigh and Sezawa modes.

The observation of relaxed wave-vector conservation for a range of scattering angles is somewhat unexpected in light of the results reported in Ref. 10 for the collinear  $(\beta=0^{\circ})$  case: there it was found that the wave-vector-

 $k_{g}$  $\beta$  $k_{g}$  $k_{g}$  $k_{g}$  $k_{g}$  $q^{+}$ 

FIG. 3. Wave-vector diagram in the plane of the grating surface illustrating the grating-relaxed Brillouin-scattering process. The grating wave vector is  $\mathbf{k}_g$ ,  $\mathbf{q}^{\pm}$  are the phonon wave vectors, and  $\mathbf{k}_{\parallel}^{\parallel}$  and  $\mathbf{k}_{\parallel}^{\parallel}$  are the parallel wave-vector components of the incident and scattered light, respectively.

FIG. 4. Polarization dependence of the Brillouin spectra for  $\beta = 90^{\circ}$ . (a) Incident polarization *H*, no analyzer. (b) Incident polarization *H*, analyzer *H*, demonstrating the suppression of the split-off modes. (c) Incident polarization *H*, analyzer *V*, demonstrating the suppression of the conventional Brillouin signal.



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FREQUENCY SHIFT (GHz)

TABLE I. Polarization characteristics of peaks observed in the Brillouin spectra for  $\beta = 90^\circ$ . The letters V and H refer to the vertical and horizontal polarizations of the light with respect to a horizontal-scattering plane. The first and second letters indicate the polarization of the incident and scattered light, respectively.

	VV	VH	HV	HH
Conventional Rayleigh and Sezawa equation (1)	yes	no	no	yes
Split-off Rayleigh and Sezawa equation (4)	yes	no	yes	no

beam. No analyzer was used because, as indicated in Sec. I, light scattered by the ripple mechanism preserves its polarization. It was, therefore, surprising to discover a strong polarization dependence when the spectrum shown in Fig. 1(b) was repeated with a horizontal and vertical polarization analyzer in the scattered beam. The HH + HV, HV, and HH spectra are shown in Fig. 4. In this notation the first and second letters indicate the polarization of the incident and scattered light, respectively. As anticipated, the conventional Rayleigh and Sezawa modes have the expected polarization-preserving dependence and do not appear in the cross-polarized spectrum. In contrast the split-off mode is completely vertically (V) polarized—appearing only in the cross-polarized



PHONON WAVE VECTOR ( 10<sup>-6</sup> m<sup>-1</sup>)

FIG. 5. Rayleigh wave velocity (squares) and Sezawa wave velocity (triangles) for phonon propagation parallel (solid symbols) and perpendicular (open symbols) to the grating grooves. There is no change within the experimental error in the Rayleigh wave velocity; however, there appears to be a small change in the Sezawa wave velocity.

spectrum—and is totally absent from the parallelpolarization spectrum.

The complete polarization dependence is even more surprising. If the incident polarization is changed from H to V, then the depolarization effect is no longer observed; all the modes appear in VV and none appears in VH. The full polarization dependence is summarized in Table I.

Before describing the role of surface plasmons in the origin of the split-off modes, a brief observation concerning the conventional Rayleigh and Sezawa modes is in order. We found that, as previously reported in Ref. 10, there was no change in the Rayleigh wave velocity for propagation parallel or perpendicular to the grooves. The velocity of the Sezawa waves does, however, show a small but consistent reduction in velocity for propagation parallel as compared to perpendicular to the grooves. This behavior is illustrated in Fig. 5, which plots the velocities of the Rayleigh and Sezawa waves versus phonon wave vector.

### IV. EFFECT OF SURFACE-PLASMON MEDIATION

The preceding results can be understood by considering the role of surface plasmons (SP's) as intermediate states in the scattering process. SP's are collective excitations of the electrons at the interface between a metal and an insulator<sup>11,12</sup> and they have been shown to enhance a number of nonlinear surface optical phenomena<sup>13,14</sup> including Brillouin scattering.<sup>10,15-17</sup> Surface plasmons on a plane surface are nonradiative; however, the relaxation of wave-vector conservation provided either by a surface corrugation, a surface acoustic wave, or by a stationary grating allows coupling of light to SP's and vice versa.

The key feature in explaining the observed polarization behavior is the fact that SP's are a transverse magnetic excitation and SP generation via grating coupling is a very sensitive function of the incident light polarization. The grating-mediated reradiation of SP's into light is expected to be similarly polarization sensitive. From Table I it can be seen that the split-off modes appear only if the analyzer is set to pass V-polarized light, i.e., only if there is a component of the E-field vector of the scattered light parallel to the vector  $k_g$ . Although, as will be discussed below, the polarization behavior is not completely understood, it is certainly suggestive of SP mediation.

The possibility of SP's being real intermediate states in the Brillouin-scattering process has a significant consequence on the spatial distribution of the scattered light collected in an actual experiment with a finite-collection aperture. To demonstrate this we reconsider the derivation of Eq. (3) in which we had implicitly assumed that all phonons with wave vectors  $\mathbf{q}^{\pm}$  would be effective in scattering light. If we now insist that SP's should be intermediate states in the scattering process, we posit the following sequence of events: (i) scattering of the incident light by a surface acoustic wave (of wave vector  $\mathbf{q}$ ) into a surface plasmon of wave vector  $\mathbf{k}_{SP}$ ,

$$\mathbf{k}_i \sin \theta_i + \mathbf{q} = \mathbf{k}_{\rm SP} , \qquad (5)$$

and (ii) reradiation of the SP with the help of a grating wave vector  $\mathbf{k}_{\sigma}$ ,

$$\mathbf{k}_{\rm SP} + \mathbf{k}_{\rm g} = \mathbf{k}_{\rm s} \sin \theta_{\rm s} \ . \tag{6}$$

The consequence of this sequence on the spatial distribution of scattered light can be visualized by redrawing the wave-vector diagram of Fig. 3 for the case of  $\beta = 90^{\circ}$  and with the stipulation that only scattering processes with SP's as intermediate states are considered. The wave-vector relationship governing this process is shown in Fig. 6.

At the single energy of the incident laser light the only allowed SP states will form a circle of radius  $k_{SP}$  in the k space of the surface. The wave-vector condition for scattering in the direction  $\mathbf{k}_s$  is shown and the phonon causing the scattering will be identical to that predicted by Eq. (3) for  $\beta = 90^{\circ}$ . This analysis assumes, however, that the collection optics measures only an infinitesimally small region in k space. In practice a real collection lens gathers light in a finite solid angle. Translating this solid angle of collection into a region of states in k space results in the area outlined by the dashed circle in Fig. 6. If SP's were not involved in the scattering process, then the Brillouin-scattered signal due to the split-off modes should be distributed evenly throughout this region of  $\mathbf{k}$ space and hence over the entire solid angle gathered by the collection lens in real space.

For the case considered in Fig. 6 in which SP's act as intermediate states, the spatial distribution is quite different. As shown in the figure the split-off-mode signal will be confined to the curved "line" in  $\mathbf{k}$  space which is an image of the segment of the surface-plasmon disper-



FIG. 6. Wave-vector diagram illustrating the SP-mediated Brillouin-scattering process. The circle of radius  $k_{\rm SP}$  is the set of possible SP states at the energy of the incident laser light. Incident light of wave vector  $\mathbf{k}_i$  is scattered by the thermal distribution of surface phonons into SP's in all directions in  $\mathbf{k}$  space. The normally nonradiative SP's can convert to scattered light with the aid of a grating wave vector  $\mathbf{k}_g$ . The Brillouin signal is confined to the curved line in  $\mathbf{k}$  space which is an image of the generated SP's translated by  $\mathbf{k}_g$ . The dashed circle represents the region in  $\mathbf{k}$  space which will be seen by a finite-collection aperture.

sion curves translated in k space by the grating wave vector  $\mathbf{k}_g$ . For the sake of clarity Fig. 6 demonstrates only translation of the upper section of the SP dispersion curve by a grating wave vector  $\mathbf{k}_g$ . By symmetry there is a  $\mathbf{k}_g$  of the opposite sign which translates the lower portion of the SP dispersion curve into the area of k space corresponding to the collection optics. This implies that the Brillouin signal due to this mechanism should be confined to a crossed pair of curved lines in k space. This restriction of the light in k space should result in a similar confinement of the signal in real space.

We could verify such a spatial restriction of the scattered light directly by experiment. In practice the wavevector-altering behavior of the surface phonons is mimicked by the natural surface roughness with the difference that scattering by the static surface roughness does not lead to a shift in frequency. This elastically scattered roughness signal is much stronger than the phonon signal, so strong, in fact, that in the scattered light gathered by the collection lens it was possible to see, albeit weakly, the crossed-line pattern predicted in the analysis above. This elastically scattered signal has the same polarization dependence as that given in Table I for the phonon signal. Furthermore, by masking out the cross pattern in the collected light it is possible to eliminate the split-off phonon signal while retaining the true Rayleigh and Sezawa signals. This proves that the split-off signal is indeed confined to the cross-line region, and verifies the correctness of the scattering sequence, described by Eqs. (5) and (6), in which SP's act as intermediate states. It is worth noting that the alternate scattering sequence described by

$$\mathbf{k}_i \sin \theta_i + \mathbf{k}_g = \mathbf{k}_{\mathrm{SP}} , \qquad (7)$$

$$\mathbf{k}_{\rm SP} + \mathbf{q} = \mathbf{k}_s \sin\theta_s \tag{8}$$

in which the incident light is grating coupled into a SP and then scattered by a phonon would not lead to the cross pattern observed, but rather to a signal uniformly distributed over the collected scattered light. Furthermore, this process can occur only if the light is incident on the sample at the appropriate angle to satisfy Eq. (7).

A further test of this scattering sequence was to consider the expected behavior for scattering configurations in which  $\beta \neq 90^{\circ}$ . In this case the split-off signal must derive from the translation of the SP dispersion curve into scattered light by a grating wave vector  $\mathbf{k}_g$ , but with  $\mathbf{k}_g$  canted at an arbitrary angle,  $\beta$ , with respect to  $\mathbf{k}_i$  and  $\mathbf{k}_s$ . The scattering process is equivalent to that shown in Fig. 3, but with the stipulation that the initial scattering process by phonons  $q^{\pm}$  carry the incident light into a SP. For  $\beta \neq 90^{\circ}$  the phonons  $q^+$  and  $q^-$  are clearly inequivalent and will lead to different frequency shifts. There should thus be a splitting in the Brillouin peaks resulting from scattering by this mechanism as the angle  $\beta$  is changed away from 90°. The spectra shown in Fig. 7, obtained with  $\beta = 90^{\circ}$  and 80°, verify that this splitting does indeed occur. The spectra were taken in HV polarization so that the conventional Brillouin peaks were suppressed; for  $\beta = 80^{\circ}$  a clear splitting is observed for both the split-off Rayleigh and Sezawa modes.

INTENSITY (Arb. Units)

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In these experiments we were clearly fortuitious in our choice of grating spacing because  $k_g$  was such that for  $\beta = 90^{\circ}$  we were able to observe the SP-mediated signal. An examination of Fig. 6 indicates that if  $k_g$  were much larger or smaller than the value illustrated in the figure, the translated section of the SP states would not have fallen within the acceptance angle of the collection optics for  $\beta = 90^{\circ}$ . To observe the SP-mediated signal from gratings with less favorable values of  $k_g$ , we would have had to change the rotation angle  $\beta$  of the grating or the incident wavelength. However, in an extreme case such as



FREQUENCY SHIFT (GHz)

FIG. 7. Brillouin spectra demonstrating the splitting of the peaks for  $\beta \neq 90^{\circ}$ . The spectra were acquired with incident polarization H and an analyzer V to eliminate the conventional Brillouin signal. (a)  $\beta = 90^{\circ}$  and (b)  $\beta = 80^{\circ}$ .

 $k_g > 2k_{SP}$  it would be impossible to observe the SPmediated scattering.

Finally the enhanced surface electromagnetic fields associated with the generation of SP's has previously been shown to enhance the Brillouin-scattering cross section,  $^{15-17}$  and the magnitude of the enhancement as a function of corrugation amplitude has been explored in a previous paper.<sup>10</sup> Although we did not perform a systematic measurement of the SP-mediated enhancement in this configuration, it can be shown from Fig. 1(b) that there is a sizable enhancement. The enhancement of the Brillouin-scattering cross section is defined as the ratio of the intensity per unit solid angle of the SP-mediated signal to that of the conventional Brillouin signal. The intensities of the conventional Rayleigh and Sezawa modes and their split-off counterparts are comparable in Fig. 1(b). However, as pointed out in Sec. IV, the split-off signal is confined to a very small area of the collection lens, whereas the conventional signal is gathered from all parts of the solid angle of collection. If the cross section is normalized for the difference in solid angle of collection, the enhancement is about 140. This estimate does not account for the different scattering wave vectors of the two signals, a correction which would tend to make the enhancement value slightly larger.

## V. CONCLUSION

We have demonstrated that the presence of a surface corrugation can dramatically alter the appearance of Brillouin spectra from surface acoustic waves on metals. The essential feature underlying all of our experimental observations is that the presence of a corrugation relaxes momentum conservation in the surface and allows coupling between light and normally nonradiative, propagating SP's. In the case of collinear q and  $k_g$ , reported previously in Ref. 10, we found that the SP-mediated effects were observed only if the angle of the incident light (or the angle of collection of the scattered light) was set to the angle for resonant SP generation. For the experiments reported here, in which the grating was oriented with  $\mathbf{k}_g$  perpendicular to the scattering plane, we found that the SP-mediated effects were present over a range of incident angles and resulted in the appearance of extra peaks in the Brillouin spectra. These new peaks were due to the two-stage scattering sequence of incident light being scattered by a surface phonon into a SP and the subsequent grating-assisted reradiation of the SP into scattered light.

The scattered light resulting from this scattering sequence was found to be strongly vertically polarized (i.e., with an E-field vector parallel to  $\mathbf{k}_g$ ) independent of the incident light polarization. While it is known that SP's are a transverse magnetic excitation, it is difficult to understand why SP's propagating in arbitrary directions in the plane of the grating should be diffracted into light with such a definite polarization. A complete calculation of the electromagnetic fields in such a scattering process is required to understand fully this feature of the experiment.

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