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Brief Reports

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Brillouin scattering from corrugated Ag films: Surface-plasmon-mediated enhancement and relaxed wave-vector conservation

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We report the investigation of Brillouin scattering from thermal surface acoustic waves on Ag-coated holographic gratings. The usual wave-vector conservation condition for Brillouin scattering is modified due to the added periodicity in the direction of phonon propagation, but this effect is observed only when surface plasmons act as an intermediate state in the scattering process. The involvement of surface plasmons results in an enhancement of the Brillouin scattering cross section which depends on the grating amplitude.

Brillouin scattering from surface phonons is the dominant phonon-scattering process in materials of high absorption in which the depth of interaction of light is restricted to a small region near the sample surface.[1] This process is governed by the conservation of wave vectors parallel to the surface,

$$|\mathbf{q}| = k_i \sin\theta_i + k_s \sin\theta_s \ , \tag{1}$$

where θ_i and θ_s are the angles that the wave vectors k_i and k_s of the incident and scattered light make with the surface normal. The frequency shift ω of the scattered light is then given by

$$\omega = v|\mathbf{q}| \ , \tag{2}$$

where v is the velocity of the scattered phonon. If an additional periodicity, such as a surface corrugation, is added in the direction of phonon propagation, the parallel-wave-vector conservation condition should, in principle, only be obeyed modulo $\mathbf{k}_g$, where $\mathbf{k}_g$ is the wave vector of the added periodicity. We show here that under conditions in which an intermediate excitation, surface plasmons (SP's) in this case, mediates the scattering process, a relaxation of wave-vector conservation is clearly observable. Moreover, the large surface electromagnetic fields associated with SP's lead to a sizable enhancement of the Brillouin signal in a similar fashion to that observed for a variety of other nonlinear optical phenomena such as second harmonic generation,[2-4] Raman scattering from adsorbates on gratings,[5,6] coherent anti-Stokes Raman scattering[7] (CARS), and surface-acoustic-wave (SAW) Brillouin scattering in an attenuated-total-reflection prism configuration.[8,9]

Holographic gratings with sinusoidal surface profiles were produced by exposing thin films of photoresist (Shipley 1470) deposited on glass slides to two interfering expanded beams from an argon-ion laser ($\lambda = 457.9$ nm, $I \approx 3.5$ mW cm^{-2}). After development, the photoresist was coated with 300 nm of Ag by magnetron sputtering. Gratings with different amplitudes were produced by varying the exposure time on the photoresist. The grating amplitude was estimated using a technique described by Pockrand[10] and the grating spacing (≈ 720 nm) determined by measuring the first-order diffraction angle and using the grating equation. Brillouin-scattering measurements were performed using a tandem Fabry-Perot interferometer in a $(5+2)$-pass configuration.[11]

SP's are a collective oscillation of the electrons at the boundary of a metal-insulator interface. For a flat metal-vacuum interface the SP dispersion relation is given by[12]

$$k_{\mathrm{SP}}=\frac{\omega}{c}\left[\frac{\epsilon}{\epsilon+1}\right]^{1/2}, \quad (3)$$

where k_{SP} is the wave vector of the SP excitation, c the velocity of light, and $\epsilon(\omega)$ the dielectric function of the metal which must satisfy $\mathrm{Re}(\epsilon)<-1$. From Eq. (3) it can be shown that k_{SP} is always greater than the wave vector of light of the same frequency. This implies that SP's are a nonradiative excitation. Thus, SP's on a flat metal surface cannot decay as light, nor can they be generated by light incident directly on the surface. One of the ways this wave-vector matching restriction governing the interaction between SP's and light can be overcome is by corrugating the metal surface, which relaxes the wave-vector conservation condition. The generation of SP's on a grating can therefore be controlled by angle tuning so as to satisfy the relation

$$k_i \sin\theta_i + n k_g = k_{\mathrm{SP}}, \quad (4)$$

where n is an integer. For our examples, $n=1$. The resonant generation of SP's is manifested by a sharp drop in the reflectivity as shown in Fig. 1 for the grating of amplitude 10.6 nm. The efficiency of conversion of light to SP's, which is a strong function of the grating amplitude, can be monitored by measuring the depth of the reflectivity minimum, the lower the minimum the greater the efficiency of conversion of light to SP's.

Figure 2 shows two Brillouin spectra, taken from the same grating as used in Fig. 1, at different angles of the incident light. Figure 2(a) was taken with the light incident at the angle for maximum SP generation, while Fig. 2(b) was taken at an angle a few degrees away from resonance where the generation of SP's is negligible. The differences between the two spectra are quite striking. In Fig. 2(b), peaks due to the Rayleigh surface wave (R) and two Sezawa modes ($S1,S2$) are observed as expected from a metallic film of this thickness. Figure 2(a), taken under the favorable condition for SP generation, shows three additional modes corresponding to the Rayleigh (R') and Sezawa ($S1',S2'$) modes at much larger frequencies. The increase in frequency of these additional modes is due to the increase in the magnitude of the scattering-phonon wave vector by k_g as described by

$$|\mathbf{q}|=k_i\sin\theta_i+k_s\sin\theta_s+k_g . \quad (5)$$

It should be noted that the intensity enhancement of the large-q phonons described by Eq. (5) is accompanied by a decrease in the intensity of the conventional Brillouin peaks ($R,S1,S2$) described by Eq. (1). The spectra shown in Fig. 2 were taken for a grating with a relatively small amplitude and hence relatively weak SP generation. Spectra from gratings of larger amplitude showed no evidence of the conventional signal when recorded in the resonant configuration.

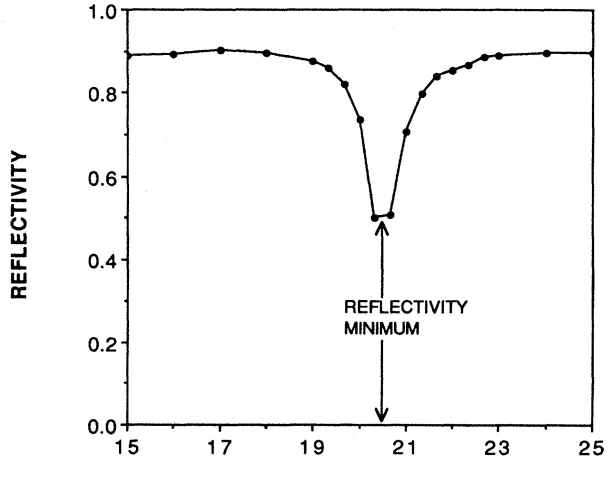

FIG. 1. Grating reflectivity as a function of angle for the grating of amplitude 10.6 nm. The most efficient SP generation occurs at the angle at which the reflectivity is a minimum.

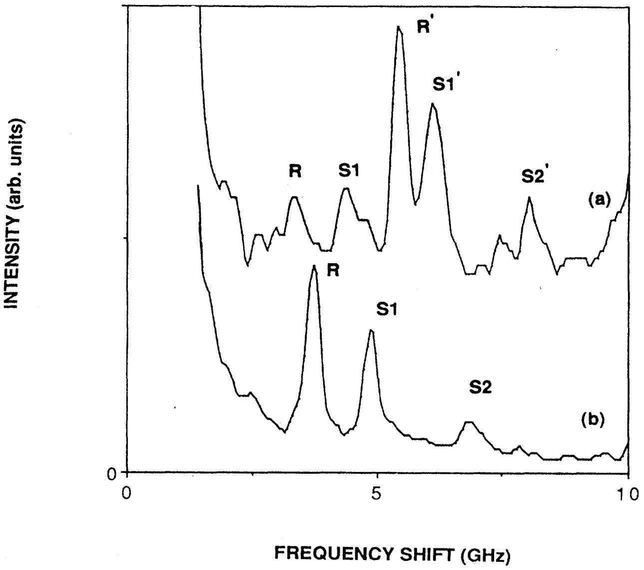

FIG. 2. Brillouin scattering spectra taken from a grating of amplitude 10.6 nm with light incident (a) at the angle for resonant SP generation and (b) a few degrees away from resonance. The peaks due to conventional Brillouin scattering by the Rayleigh and Sezawa modes are indicated by R, $S1$, and $S2$, while the equivalent SP-mediated Brillouin peaks are marked R', $S1'$, and $S2'$.

In Fig. 3 the values of the SAW velocities are plotted as a function of phonon wave vector $\mathbf{q}$. The lowest velocity mode is due to the Rayleigh surface wave while those of higher velocity are due to the Sezawa modes. The solid symbols at large $\mathbf{q}$ are due to Brillouin scattering with the light incident at the coupling angle for resonant generation of SP's. The arrow on the figure indicates the concomitant increase in the wave vector of the scattering phonon by a factor k_g.

The value of the SP-mediated enhancement in the

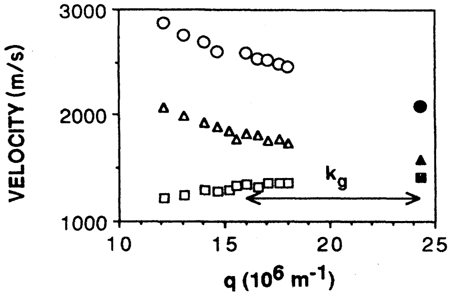

FIG. 3. Surface-acoustic-wave velocity measured as a function of phonon wave vector. Squares, triangles, and circles refer to the Rayleigh, first Sezawa, and second Sezawa modes, respectively. The open symbols are due to conventional Brillouin scattering, while the solid symbols indicate the SP-mediated signal with phonon wave vector increased by the addition of k_g.

TABLE I. Grating parameters and measured Brillouin enhancement values.

Exposure Time (sec)	Grating amplitude (nm)	Reflectivity minimum	Enhancement factor
10	10.6	0.5	8
20	23.8	0.15	23
25	30.4	0.04	29
30	33.0	0.06	26

SAW Brillouin scattering was determined by measuring the intensity of the surface Brillouin signal at resonance and comparing it to that measured a few degrees away from resonance. The enhancement thus determined requires two corrections.

(i) Different incident powers had to be used in acquiring the two spectra because, when SP's were resonantly produced, the threshold for damage to the surface was of the order of 80 mW with a focused spot of ~100 μm diameter. Most of the SP-resonance spectra were therefore acquired with incident power levels of about 30 mW. In contrast, to achieve well-defined spectra in the off-resonance cases, it was necessary to increase the power to between 250 and 400 mW.

(ii) Because phonons of different wave vectors were probed in the resonant and nonresonant cases, and since the intensity of the Brillouin scattered signal is inversely proportional to the square of the frequency shift,[1] we have incorporated a correction factor equal to the square of the ratio of the measured frequency shifts.

The values of the measured grating parameters and the final enhancement values are listed in Table I for each of the four gratings. As expected, the maximum enhancement is achieved for the grating with the deepest reflectivity dip where SP generation is strongest. The estimated error in the enhancement values is ~±20%, which is only slightly lower than the errors encountered using prism generation of SP's.[8,9] However, the measurements themselves are considerably easier in the grating configuration because the change between the enhanced and unenhanced configurations by angle tuning requires little optical realignment and because spurious signals from bulk Brillouin scattering in the coupling prism are eliminated.

The phonon velocities measured on gratings with different amplitudes were the same within experimental error (±2%) as that measured on a flat surface for the range of grating amplitudes studied in these experiments. This result indicates that the corrugation does not strongly affect the propagation of surface waves. We note, however, that the largest effect is expected[13] at $q \sim k_g/2 = 4\times10^6$ m^{-1} which, as seen in Fig. 3, is not the region we investigated.

In summary, we observe a relaxation of the wave-vector conservation requirements for Brillouin scattering due to the added periodicity in the direction of phonon propagation but *only* when the incident light is coupled to SP's. The involvement of SP's in the scattering process also leads to an enhancement in the Brillouin scattering cross section. Although other nonlinear optical phenomena have been observed via grating coupled SP's, Brillouin scattering is unique in that, in addition to an enhancement in the scattering cross section, the requirements of wave-vector conservation lead to a shift in the frequency of the scattered light when SP's are involved. This separation in frequency is a clear demonstration of SP mediation and permits a direct distinction between the enhanced and unenhanced signals.

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