Second harmonic generation due to the excitation of oppositely propagating surface polaritons on a quartz surface

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The generation of the second harmonic (SHG) ~5μm in a direction perpendicular to the surface of the sample is observed as a result of the excitation of oppositely propagating surface polaritons on quartz with a diffraction grating (18.2 μm) by ~10 μm infrared radiation. The SHG is maximum in the region of the 10-cm^{-1} gap, produced by the diffraction grating, on the dispersion curve for quartz surface polaritons. © *1995 American Institute of Physics.*

Surface polaritons (SPs) are excitations whose electric field is maximum at the interface and decays exponentially away from the interface.[1] This makes surface polaritons a sensitive indicator of the state of the surface, and the amplification of the field accompanying excitation of surface polaritons should make nonlinear optical processes at the surface more efficient.[2]

Visible-range second-harmonic generation (SHG) which accompanies the excitation of surface plasmons-polaritons on a metal, was observed in Refs. 2 and 3. A scheme in which oppositely propagating surface plasmons-polaritons are simultaneously on a diffraction grating was used in Ref. 3. We employed the same scheme to obtain middle-infrared-range SHG on a quartz surface with excitation of oppositely propagating surface phonons-polaritons of quartz.

In quartz the highest-frequency E modes fall within the tuning range of a CO_2 laser, and the propagation of the surface polaritons on the surface of the quartz in this region has been well studied.[4–6] Surface polaritons are nonradiative oscillations. Their wave vector exceeds the wave vector of radiation with the same frequency. We employed lattice conversion of surface polaritons to excite surface polaritons on quartz.[1]

The excitation of surface polaritons on a diffraction grating occurs when the wave vector k_{sp} of the surface polaritons is equal to the sum of the projection of the wave vector of the radiation and a vector that is a multiple of a reciprocal lattice vector. If the grating lines are perpendicular to the plane of incidence of the radiation, then this corresponds to

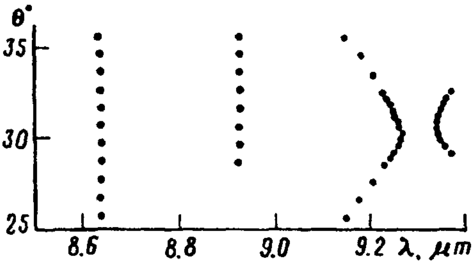

FIG. 1. Frequency dependence of the minima in the reflection spectra of one of the quartz samples with a diffraction grating (the lattice period is equal to 18.2 μm; the lines are etched to a depth of 0.9 μm) plotted as a function of the angle of incidence of the radiation.

$$k_{sp}=\frac{2\pi}{\lambda}\sin\theta+\frac{2\pi p}{a},\quad p=0,\ \pm 1,\ \pm 2,\ \pm 3,\ldots,$$

where λ is the radiation wavelength, θ is the angle of incidence of the radiation, a is the lattice constant, and p is the diffraction order. For angle of incidence of the radiation close to 30°, the condition for simultaneous excitation of surface polaritons propagating in the direction of propagation of the radiation ($p=+1$) and in the opposite direction ($p=-3$) can be satisfied.

The diffraction gratings were deposited on a quartz surface by the method of photolithography followed by ion etching. The optimal depth of the gratings was determined so as to maximize the excitation of surface polaritons on the quartz surface and was equal to 0.9 μm. The period of the gratings was chosen to be 18.2 μm for excitation of quartz surface polaritons near 9.3 μm. The grating lines were parallel to the quartz optical axis, lying in the plane of the sample (in this case the anisotropy of the optical constants of quartz has no effect on the surface-polaritons spectra).

The infrared-reflection spectra of the quartz samples with deposited gratings at different angles of incidence of the radiation were investigated with a Michelson 110 Fourier spectrometer (BOMEM). When surface polaritons are excited, minima are observed in the reflection spectra for different values of p. The frequency dependence of their position on the angle of incidence gives the dispersion of the surface polaritons (Fig. 1). One can see that as a result of the interaction of the surface polaritons, the dispersion curves split at the point where they cross ($\lambda=9.3$ μm; $\theta=30.6°$).

As reported in Ref. 3 , the strong SHG signal on silver diffraction gratings was observed at frequencies falling in the gap in the dispersion curve of the surface polaritons at $\lambda=850$ nm. We decided to perform similar experiments on quartz, using excitation of surface phonons-polaritons.

Radiation from a free-electron laser FELIX[7] with the following parameters was used to observe SHG: macropulse duration 4.2 ms, repetition frequency 5 Hz, micropulse duration 1.3 ps, repetition frequency 1 GHz, and peak power per micropulse 5 MW. The wavelength of the radiation varied from 8.5 to 9.8 μm with a step of 0.05 μm. The spectral width of the radiation line was equal to 0.1 μm, P-polarized radiation of the

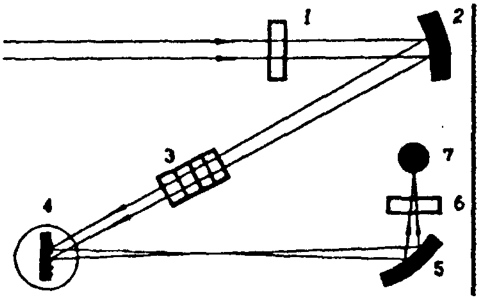

FIG. 2. Diagram of the experimental apparatus for observing SHG. 1 — Filter cutting off radiation with wavelengths shorter than 7 μm; 2 — spherical mirror (f = 1 m); 3 — grid attenuator, making it possible to change the intensity of the incident radiation by 3, 5, and 10 dB; 4 — sample (quartz with a diffraction grating); 5 — parabolic mirror; 6 — 0.4-cm-thick sapphire plate which absorbs radiation with $\lambda = 9.3$ μm but transmits radiation with $\lambda = 4.65$ μm; 7 — nitrogen-cooled radiation detector (HgCdTe).

beam (Fig. 2) was collected with a spherical mirror 2 (f = 1 m) on the sample 4 located 30 cm from the focus. In this case the diameter of the radiation spot on the sample was equal to 0.23 cm. The sample was placed on a table of a theodolite with a angular division of 0.05°. The angle of incidence θ of the radiation was set at 30.6° (the angular width of the radiation beam was equal to 0.6°). A grid attenuator 3 placed in front of the sample made it possible to change the intensity of the incident radiation by 3, 5, and 10 dB, and a filter 1 absorbed radiation with wavelengths shorter than 7 μm (the transmission of the filter at 5 μm was less than 0.1%). The radiation propagating from the sample in the direction of the normal (second harmonic) was focused by a parabolic mirror 5 on the liquid-nitrogen-cooled radiation detector 7. A 0.4-cm-thick sapphire plate 6, which absorbed radiation with $\lambda = 9.3$ μm (the transmission coefficient for 10-μm radiation is equal to less than 0.1%) but transmits radiation with $\lambda < 5$ μm, was placed in front of the detector.

Figure 3 shows the shape of the macropulse of radiation incident on the sample (top

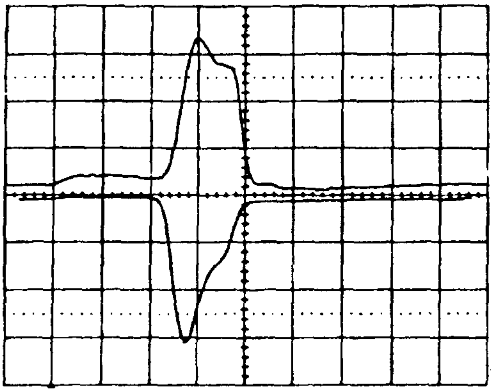

FIG. 3. Shape of the macropulse of radiation incident on the sample (top curve) and shape of the signal pulse from the detector (bottom curve) corresponding to SHG on the quartz surface.

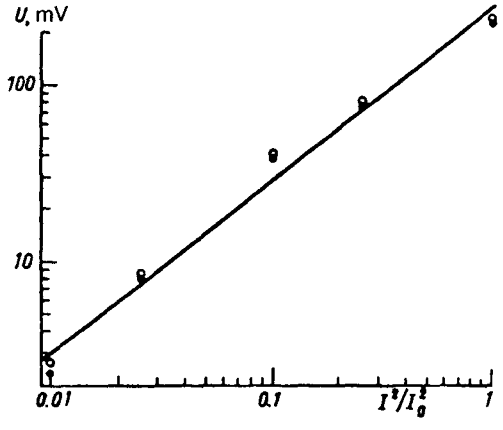

FIG. 4. SHG intensity as a function of the squared intensity of the incident radiation ($\lambda=9.3$ μm). Dots — experimental values; solid line — linear approximation corresponding to a quadratic dependence of the SHG on the intensity of the incident radiation.

curve) and the shape of the signal pulse of the detector corresponding to SHG on the quartz surface (bottom curve). The distortion of the pulse shape is characteristic of nonlinear processes. It is due to the high radiation power density at the start of the macropulse (because of the shorter duration of the micropulses). In our experiment the micropulse duration at the start of the macropulse was equal to 1.3 ps.

We measured the dependence of the detector signal on the intensity of the incident radiation, which was varied with a grid attenuator (3, Fig. 2). The dependence of the SHG intensity on the squared intensity of the incident radiation (wavelength 9.3 μm) is shown in Fig. 4: dots — experimental values, solid straight line — linear approximation corresponding to a quadratic dependence of SHG on the intensity of the incident radiation. The observation of the quadratic dependence of the signal intensity on the intensity of the incident radiation confirms SHG on this sample.

To check the fact that SHG is associated with surface polaritons excited on the quartz surface with the help of the gratings, we performed the following measurements:

1) We changed the orientation of the sample: We set the grating lines parallel to the plane of incidence of the radiation. In this case the conditions for excitation of surface polaritons are not satisfied. The signal strength drops to the noise level;

2) We displaced the sample so that the radiation was incident on the smooth surface of the quartz. We did not excite the surface polaritons in this case. The signal strength dropped to the noise level.

Surface polaritons on quartz exist in a narrow spectral range, and correspondingly SHG should be observed in the same spectral range. The frequency dependence of the SHG signal was measured at the optimum angle of incidence of the radiation on the sample of 30.6°. The plot is shown in Fig. 5 (bottom curve). The maximum signal strength occurs at a wavelength of 9.3 μm, in good agreement with the position of the

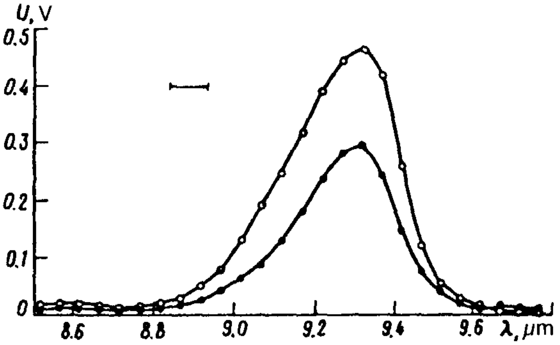

FIG. 5. Frequency dependence of the SHG signal on the clean surface of the quartz sample with a diffraction grating (period 18.2 μm; etch depth of the lines 0.9 μm) — filled circles and coated with a 200-Å-thick insulating — open circles. The solid lines are drawn through the experimental points. The bar in the upper left-hand corner represents the spectral linewidth of the FELIX radiation.

gap in the dispersion curve of the surface polaritons for this sample (Fig. 1). The SHG excitation efficiency was equal to 1.2×10^{-9}.

When a transparent insulating film is present on the surface, the intensity of the surface polaritons is more highly concentrated near the surface of the sample.[1] It could therefore be expected that the SHG intensity should increase in the presence of such a film. A 200-Å-thick insulating film was deposited on the sample by the Langmuir–Blodgett method. One could see in the reflection spectra that this resulted in a 30% increase of the absorption in the bands corresponding to excitation of surface polaritons. The frequency dependence of the SHG signal could also be measured on the same sample. This dependence is shown in Fig. 5 by the top curve. The SHG signal strength also increased. This result is an additional confirmation of the fact that the observed SHG is due to surface polaritons excited on the quartz surface.

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