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SPECTROSCOPY

PHASE SPECTROSCOPY OF SURFACE ELECTROMAGNETIC WAVES

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Abstract—Surface EM waves (SEMW) have been found to be a sensitive indicator of the state of a surface. By measuring the amplitude of a SEMW travelling along a specimen one may obtain information on the surface roughness, adsorbates and oxide films. Phase spectroscopy which determines the phase change of a surface wave propagating along the specimen (effective refractive index for SEMW) provides a new analytical tool and extends the range of objects that can be investigated. For two wavefronts launched at two points on the surface carrying the surface wave (these may be the exciting element and specimen's edge), computer assisted analysis of intensity distribution determines the apparent refractive index and the respective absorption coefficient. Phase spectroscopy of surface waves was used to study metals with natural oxide films in two modes: (1) laser excitation of surface waves (CO_2 laser light tunable in the range 930–1088 cm^{-1}) and (2) wideband excitation (700–2500 cm^{-1}) coupled with a Fourier spectrometer. Laser excitation was also used to investigate dielectrics (crystalling quartz with metal films) and high-temperature superconductors $YBa_2Cu_3O_{7-\delta}$ (ceramics, single crystals and films on strontium titanate). Optical constants were evaluated for all the materials studied.

The amplitude of the field of surface EM waves (SEMW) is greatest at the interface between two media and falls off away from this boundary. The distribution of SEMWs along the interface is described in terms of the effective refractive index of SEMWs [1]

$$\chi_x = \frac{k_x c}{\omega}, \tag{1}$$

where

k_x is the wave vector of SEMW

ω is the angular frequency.

At the interface between a vacuum and a medium of dielectric permittivity ε, the effective refractive index is given by the formula [1]

$$\chi_x = \sqrt{\frac{\varepsilon}{\varepsilon + 1}}. \tag{2}$$

For a complex permittivity, the quantity χ_x is also complex valued. Its imaginary part represents the spatial attenuation of a SEMW and can be determined by measuring how the intensity varies with the distance travelled by this wave (amplitude spectroscopy). The real part of the effective refractive index (phase spectroscopy of SEMW) can be determined by way of interference measurements [2, 3].

Figure 1 outlines the idea of such an experiment with a prism for launching a surface EM wave. In the gap between the specimen and the screen, the wave gets partially transformed into bulk radiation. The remaining portion of the surface wave also becomes bulk radiation at the edge of the specimen. These two beams interfere with one another, and if there is an interference maximum or minimum at a point z [3]

$$a \operatorname{Re} \chi_x + \sqrt{b^2 + z^2} - \sqrt{(a+b)^2 + z^2} = (m + \Delta)/\nu \tag{3}$$

where

a is the distance from screen to specimen edge,

b is the distance from specimen edge to observation plane,

m is an integer for maxima, or half-integer for minima,

$\nu = \omega/2\pi c$ is the linear frequency (wavenumber).

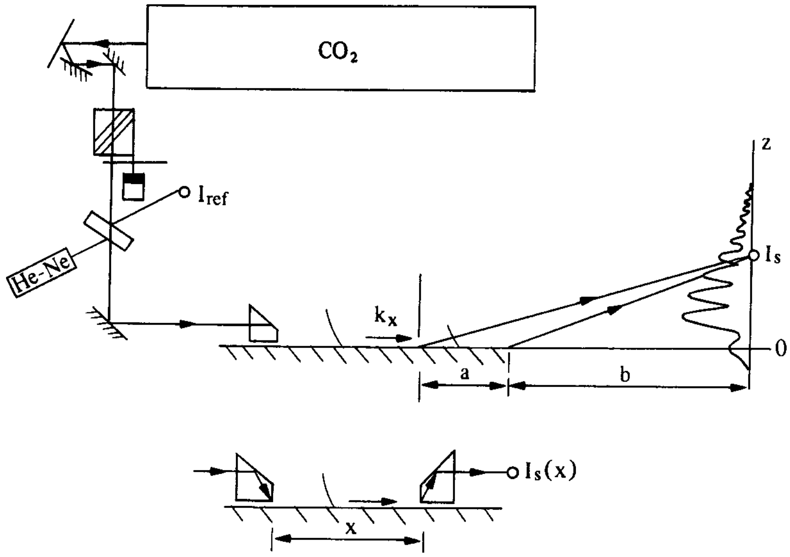

Fig. 1. Experimental arrangement for interference measurements (top) and for double prism measurements of surface wave absorption (bottom).

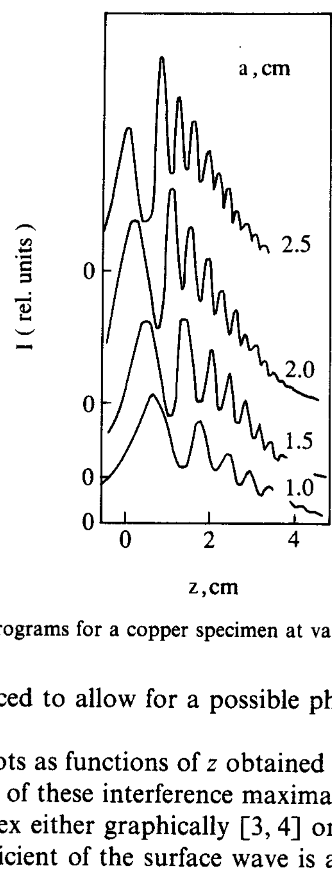

Fig. 2. Interferograms for a copper specimen at various values of a.

The quantity Δ has been introduced to allow for a possible phase shift between a bulk wave and the SEMW.

Figure 2 shows the intensity plots as functions of z obtained in moving the pyroelectric detector at a constant speed. The position of these interference maxima may be used to determine the real part of the effective refractive index either graphically [3, 4] or by least squares (by Eq. (3)) on a computer. If the absorption coefficient of the surface wave is also known (from measurements of intensity of the SEMW versus distance), then the dielectric permittivity of the specimen can be determined. Zhizhin *et al.* [4] and Silin [5] have reported such measurements for copper and vanadium.

The equation of dispersion of SEMWs (2) at a metallic surface becomes more complicated when this surface is covered by thin films, for χ_x depends on the properties and thickness of the film. Solving the inverse problem one may determine the parameters of the film by the measured values of the real and imaginary parts of the SEMW effective refractive index. Voronov and co-workers

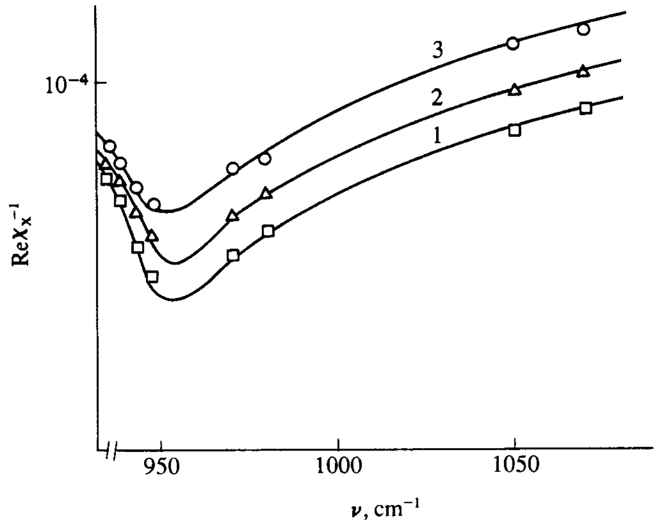

Fig. 3. Frequency dependences of the real part of the effective index of refraction of SEMW in aluminium coated with thermally grown oxide films.

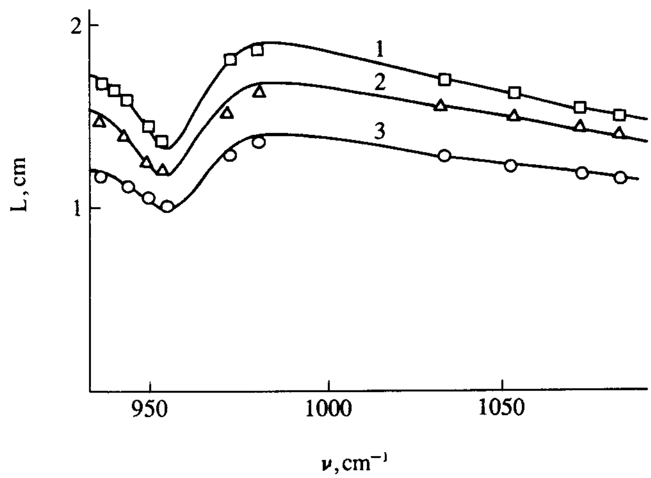

Fig. 4. Frequency dependences of SEMW path length in aluminium with thermally grown oxide films.

[6, 7] have studied oxide films on copper and aluminium. Figures 3 and 4 plot the frequency dependencies of $\mathrm{Re}(\chi_x - 1)$ and SEMW path length $L = 1/4\pi\nu\ \mathrm{Im}\ \chi_x$ for the natural oxide film on aluminium (curve 1) and for oxides formed after heating the specimen at 250°C for 5 min (curve 2) and at 370°C for 20 min (curve 3). Using the oxide film thickness as the sole variable parameter it was calculated that the thickness increased by a factor of 2.5 after the maximum heat treatment.

In recording interference patterns, scanning by moving the detector up the z-axis (Fig. 1) is convenient only when studying good conductors. For other materials, as the absorption of the surface wave increases, the distance a at which satisfactory interference patterns can be obtained decreases. This results in wider spaced extrema. On the other hand, the lower localization of the SEMW field above the specimen causes the radiation patterns of the interfering bulk waves to expand. Angular scanning with the detector moving through the arc of a circle is more convenient in this situation. The layout of the modified experiment is illustrated in Fig. 5. The surface wave is launched at an aperture between the surface of the specimen and the screen edge [8].

In the geometry of this experiment, if $a \sim m/\nu$ is much less than the radius of the arc through which the detector scans, then

$$\chi_x' \approx \cos\theta_m + m/\nu a \tag{4}$$

where θ_m stand for the angular positions of interference extrema.

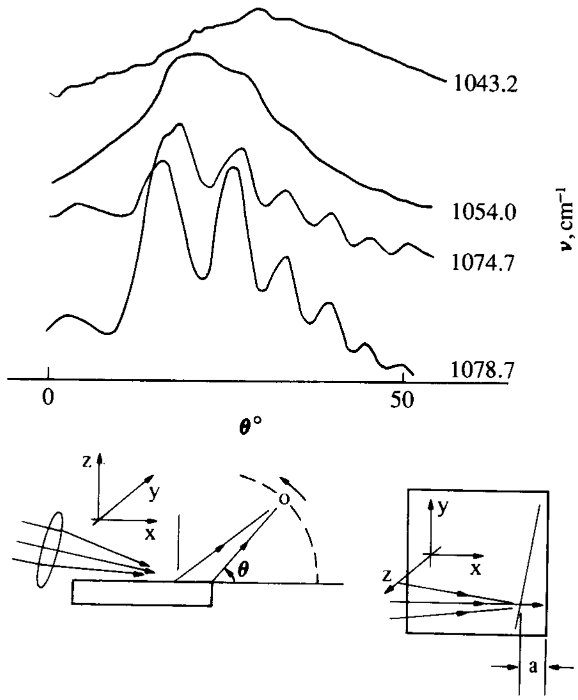

Fig. 5. Experimental arrangement for angular scanning. Interferograms for crystalline quartz at various frequencies (top).

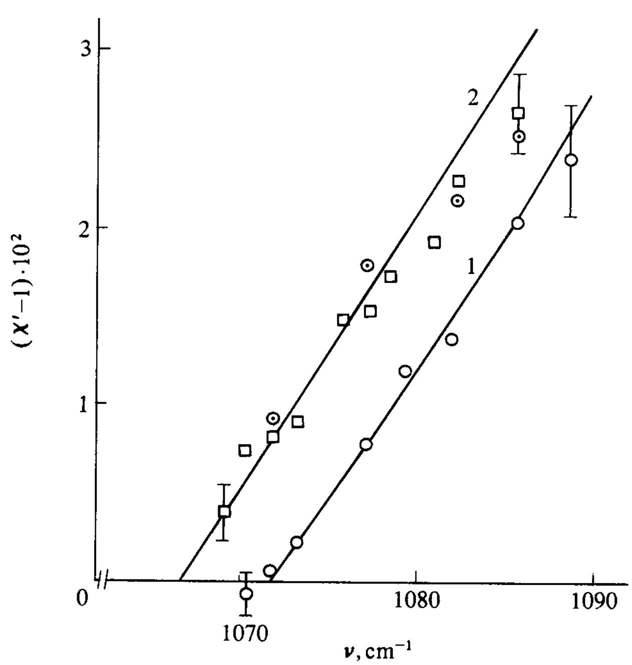

Fig. 6. Frequency plots of the real part of the SEMW refractive index for variously oriented crystalline quartz: (1) for optical axis, C, parallel to x; (2)for C parallel to y and C parallel to z. For the orientation of the coordinate axes refer to Fig. 5.

When the absorption of surface waves is strong, its measurement by ordinary methods becomes a complicated problem [5, 9]. On the other hand, the distribution of intensity in the interferogram contains information about the SEMW absorption coefficient. Voronov [7] gives the formula

$$\frac{a}{2L} + A(\theta) = \ln\left(\frac{\sqrt{I_{\max}} + \sqrt{I_{\min}}}{\sqrt{I_{\max}} - \sqrt{I_{\min}}}\right) \qquad (5)$$

where I_{max} and I_{min} are determined by the maxima and minima of the interferogram, and $A(\theta)$ is independent of a. By measuring interferograms for a number of distances a and using Eq. (5) one may determine the path length of the surface wave, i.e. evaluate both real and imaginary parts of χ_x.

Figure 6 shows the frequency dependences of the real part of the effective refractive index (dispersion of SEMW) of crystalline quartz for different direction of SEMW propagation with respect to the optical axis (anisotropy) [10].

Deposition of thin films onto the surface of the quartz specimen changed the dispersion. Films with positive dielectric permittivity increase the value of Re χ_x, and films with negative permittivity (of metal) decreased this value [11].

For the high-temperature superconductors $YBa_2Cu_3O_{7-\delta}$, the room temperature conductivity is about two orders of magnitude lower than for metals of high conductivity. Therefore the attenuation of the surface wave on these materials is high and in studying various specimens of them we resorted to the experimental arrangement shown in Fig. 5. We investigated ceramic specimens [12], single crystals [13] and films deposited on strontium titanate. For single crystals in the 10-μm range, we obtained the complex permittivity $\varepsilon \approx -45 + 90i$.

The aforementioned results were obtained by means of a CO_2 laser tunable in the range 930–1080 cm^{-1}. Chesters *et al.* [14] employed a Fourier spectrometer to perform interference measurements for silver films in a wide spectral range (700–2500 cm^{-1}).

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