and $3p\,^2P_{3/2}$ via other possible intermediate states are orders of magnitude weaker because of the smaller oscillator strength and much larger values of Δ.

Although the Raman peaks we have observed are small, saturation or near saturation is possible when the Rabi frequency Ω is *not* small compared with Δ; for instance, spontaneous Raman processes in excited states may play a significant role in opening up pathways for multiple-photon absorption in molecular photolysis, despite anharmonicity in the vibrational spectrum. This may help to explain the recent success in isotope-separation attempts using CO_2 lasers.[7]

Finally we remark that excited-state Raman processes may pose difficulties in the selective laser excitation of individual Rydberg states. Consider the excitation of a high Rydberg state (n,l), with $l=0$ or 2, from the $3p$ state of sodium. It is easy to show that the state $(n+1,l)$ would acquire, through the Raman process, a steady-state population relative to that of (n,l) which is proportional to n^3I, where I is the laser intensity. Of course, the significance of this effect would depend on laser intensity, pulse duration, and competing processes such as photoionization.

We are happy to acknowledge the fact that the apparatus was developed by M. Lambropoulos and S. Moody, who also rendered a great deal of practical assistance in this work. A Szöke was invaluable in helping us make theoretical estimates.

*Work supported in part by the National Science Foundation under Grant No. MPS72-05167.

†Joint Institute in Laboratory Astrophysics Visiting Fellow, 1975-1976, on sabbatical leave from the University of Massachusetts, Amherst, Mass. 01002.

‡Joint Institute for Laboratory Astrophysics Visiting Fellow, 1975-1976, on sabbatical leave from the University of Connecticut, Storrs, Conn. 06268.

[1]M. Lambropoulos *et al.*, Phys. Rev. Lett. 35, 159 (1975); S. E. Moody, Ph.D. thesis, University of Colorado, 1975 (unpublished).

[2]See, for example, D. Cotter *et al.*, Opt. Commun. 15, 143 (1975); J. Carlsten and P. Dunn, Opt. Commun. 14, 8 (1975); S. Barak and S. Yatsiv, Phys. Rev. A 3, 382 (1971); P. Sorokin *et al.*, Appl. Phys. Lett. 10, 44 (1967).

[3]A. Flusberg, R. A. Weingarten, and S. R. Hartmann, Phys. Lett. 43A, 433 (1973).

[4]Stark-mixing enhancement of a quadrupole transition has been seen before in cesium using fluorescence detection: P. Zimmerman *et al.*, Opt. Commun. 12, 198 (1974).

[5]E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge Univ. Press, Cambridge, England, 1951), p. 133; and R. H. Garstang, private communication.

[6]W. Heitler, *The Quantum Theory of Radiation* (Oxford Univ. Press, London, 1954), 3rd ed., pp. 190–192.

[7]See, for example, R. V. Ambartzumian *et al.*, in *Laser Spectroscopy*, edited by S. Haroche *et al.* (Springer, Berlin, 1975), pp. 121–131; and C. P. Robinson, *ibid.*, pp. 275–295.

Excitation of Surface Electromagnetic Waves in Attenuated Total-Reflection Prism Configurations*

W. P. Chen, G. Ritchie, and E. Burstein

Physics Department and Laboratory for Research in the Structure of Matter, University of Pennsylvania, Philadelphia, Pennsylvania 19174

(Received 16 August 1976)

We have carried out theoretical and experimental studies of the excitation of surface electromagnetic waves by volume electromagnetic radiation of finite beam width. The excited leaky surface electromagnetic waves are found to build up along the excitation region and to propagate as free damped waves beyond. Correspondingly, the spatial dependence of the reflected intensity is found to exhibit a two-peak interference structure at the leading edge of the excitation region followed by an exponential decay beyond.

An attenuated total-reflection (ATR) spectroscopy of metal surfaces has evolved in recent years,[1-3] which makes use of the resonant excitation of surface electromagnetic (SEM) waves by linear coupling with volume electromagnetic (VEM) waves in either an Otto configuration (i.e., prism-vacuum-metal) or a Kretschmann configuration (i.e., prism-metal-film-vacuum). We note, however, that in previous theoretical formulations of these phenomena, which have been carried out thus far,[3-5] the "incident" and "reflected" VEM waves in the prism medium have been assumed to be plane waves. This assumption is a reasonable one for ATR experiments carried out at frequencies in the visible range where the propagation length L of the "leaky" SEM waves of the three-media configuration (of the order of microns) is very much smaller than the "lateral" beam width w of the incident VEM waves. The assumption of plane waves is, however, not applicable at frequencies in the infrared where L (of the order of millimeters or more)[6-8] is comparable to or larger than w.

In this Letter, we present results of theoretical and experimental studies of the excitation of SEM waves when the incident VEM radiation has a finite beam width. We find that the spatial dependence of the "reflected" intensity exhibits a double-peak structure which is due to the interference between the "mirror"-reflected VEM radiation and the VEM radiation that is emitted by the *driven* leaky SEM waves in the region of excitation, and an exponential decay beyond the region of excitation, which is due to the "radiation" emitted by *free* leaky SEM waves. We also find that, when the angle of incidence is varied, the reflected intensity from the central part of the excitation region exhibits ATR minima, while the radiated intensity beyond the excitation region exhibits peaks.

The theoretical formulation used is similar to the one used by Tamir and Bertoni[9] to analyze the so-called "lateral displacement of optical beams" at multilayer configurations involving leaky waves. Here we specialize to the case of leaky SEM waves in an ATR prism configuration. We limit our discussion to the leaky SEM waves in an Otto prism (medium 3)–vacuum (medium 2)–metal (medium 1) configuration. The extension to a Kretschmann configuration and to prism configurations involving surface active media other than metals is straightforward and, with respect to the excitation of leaky SEM waves, one finds essentially the same phenomena.

Consider an incident beam of TM-polarized VEM radiation within the prism having a lateral beam width along the prism-vacuum interface w very much larger than either the wavelength (i.e., diffraction effects can be neglected) or the thickness d of the vacuum between the prism and the metal, but comparable to or smaller than the propagation length of the leaky SEM waves, and a "reflected" beam whose lateral beam width is determined by a slit placed in front of the detector. The beam is assumed to be essentially unlimited in the direction normal to the plane of incidence. The latter is taken to be in the xz plane; and the normal to the interfaces is taken to be along z.

At the prism-vacuum (medium 3–medium 2) interface, taken to be at $z=0$, the components of the electric field, parallel to and normal to the interface, within the prism are given by

$$E_x^{i}(\theta_i, x, z=0) = F(x)\exp(ik_x^{0}x)\cos\theta_i; \quad \frac{E_z^{i}(\theta_i, x, z=0)}{E_x^{i}(\theta_i, x, z=0)} = \tan\theta_i, \tag{1}$$

where $F(x)$ is the lateral form factor of the incident VEM beam at $z=0$, chosen for simplicity to be symmetrical with respect to its center at $x=0$; θ_i is the angle of incidence of the central ray of the incident beam; $k_x^{0} = (\omega\epsilon_3^{1/2}/c)\sin\theta_i$, and a time dependence $\exp(-i\omega t)$ is implied.

$E_x^{i}(\theta_i, x, z=0)$ can be expressed as a Fourier integral over plane waves as follows:

$$E_x^{i} = E_x^{i}(\theta_i, x, z=0) = \int_{-\infty}^{\infty}\Phi(k_x)\exp(ik_x x)dk_x = \int_{-\infty}^{\infty}E_x(k_x)\,dk_x, \tag{2}$$

where

$$\Phi(k_x) = 2\pi^{-1}\int_{-\infty}^{\infty}E_x(\theta_i, x, z=0)\exp(-ik_x x)\,dx. \tag{3}$$

Since $E_z(x,z)$ can be related to $E_x(x,z)$ by Maxwell equations in each medium, we need only, in what follows, focus our attention on $E_x(x,z)$.

The electric field of the EM radiation which is "reflected" back into the prism and the electric field of the EM radiation which is "transmitted" into the metal at the metal-vacuum interface are given by

$$E_x^{r} = E_x^{r}(\theta_i, x, z=0) = \int_{-\infty}^{\infty}\tilde{r}(k_x)E_x(k_x)\,dk_x, \tag{4}$$

$$E_x^{t} = E_x^{t}(\theta_i, x, z=-d) = \int_{-\infty}^{\infty}\tilde{t}(k_x)E_x(k_x)\,dk_x, \tag{5}$$

where $\tilde{r}(k_x)$ and $\tilde{t}(k_x)$ are the complex reflectance and complex transmittance, respectively, of the three-medium configuration, for an incident plane wave in the prism, which are readily shown to be given by

$$\tilde{r}(k_x) = \frac{(\epsilon_2 k_{1z} + \epsilon_1 k_{2z})(\epsilon_3 k_{2z} - \epsilon_2 k_{3z}) + (\epsilon_z k_{1z} - \epsilon_1 k_{2z})(\epsilon_3 k_{2z} + \epsilon_2 k_{3z}) \exp(i2k_{2z}d)}{(\epsilon_2 k_{1z} + \epsilon_1 k_{2z})(\epsilon_3 k_{2z} + \epsilon_2 k_{3z}) + (\epsilon_2 k_{1z} - \epsilon_1 k_{2z})(\epsilon_3 k_{2z} - \epsilon_2 k_{3z})\exp(i2k_{2z}d)} = \frac{N}{D}, \tag{6}$$

$$\tilde{t}(k_x) = \frac{4\epsilon_2\epsilon_3 k_{1z} k_{2z} \exp(ik_{2z}d)}{D}, \tag{7}$$

where ϵ_j is the dielectric constant of medium j ($j = 1$, 2, and 3) and $k_{jz} = [(\omega^2/c^2)\epsilon_j - k_x^2]^{1/2}$ is the z component of the wave vector of the EM fields in medium j. The roots of D correspond to a set of "leaky" modes. The modes for which k_{2z} and k_{1z} are predominantly imaginary are the "leaky" SEM modes which are linearly coupled to and, therefore, "radiate" VEM radiation in the prism.

We can express $\tilde{r}(k_x)$ in terms of the roots $\tilde{k}_s = k_s' + ik_s''$ of D, corresponding to the complex propagation wave vectors of the "leaky" SEM waves, when k_x is close to k_s', as follows:

$$\tilde{r}(k_x) = A + B(k_x - \tilde{k}_s)^{-1}; \quad \tilde{t}(k_x) = C(k_x - \tilde{k}_s)^{-1}, \tag{8}$$

where $A \cong (N'/D')|_{\tilde{k}_s}$, $B \cong (N/D')|_{\tilde{k}_s}$, $C = [4\epsilon_3\epsilon_2 k_{2z} k_{1z} \exp(ik_{2z}d)/D']|_{\tilde{k}_s}$, D' and N' are the derivatives of D and N with respect to k_x evaluated at $k_x = \tilde{k}_s$. On substituting these expressions into Eqs. (4) and (5) and applying convolution theory, we obtain

$$E_x^r = AE_x^i + iBE_x^s, \quad E_x^t = iCE_x^s, \tag{9}$$

$$E_x^s = \exp(i\tilde{k}_s x) \int_{-\infty}^{x} E_x^i(\theta_i, x', z=0) \exp(-i\tilde{k}_s x')\, dx'. \tag{10}$$

The first term of E_x^r represents the "mirror reflection" of the incident beam, in the absence of SEM wave excitation, which is due primarily to the reflection at the prism-vacuum interface. The second term of E_x^r represents the field of the VEM waves "radiated" by the generated SEM waves. The constant B is predominantly imaginary and positive. The first and second terms are therefore out of phase with each other. Furthermore, because of the difference in the spatial distribution of the mirror reflected and the radiated components, the "reflected intensity" will generally exhibit a double-peak spatial structure. The appearance of a second peak has been attributed to the "lateral displacement of the reflected beam."[9] However, unlike the Goos-Hänchen effect in which there is an actual lateral displacement of the reflected beam, the appearance of the second peak is due primarily to a destructive interference between the mirror-reflected and radiated fields, which have different spatial distributions. The constant C is predominantly imaginary and negative. Thus, E_x^t has essentially the same phase as E_x^i.

We note that the integral of Eq. (10) is negligible for $x < w/2$, where $E_x^i(\theta_i, x, z=0) \cong 0$, and is equal to a constant S for $x > w/2$. Thus for $x > w/2$, i.e., for x beyond the excitation region, the expressions for E_x^r and E_x^t reduce to $E_x^r(\theta_i, x, z=0) \cong iBS\exp(i\tilde{k}_s x)$ and $E_x^t(\theta_i, x, z=-d) \cong iCS\exp(i\tilde{k}_s x)$. These fields represent *free* "leaky" SEM waves which have a propagation length $L = 1/(2k_s'')$ and which radiate energy within the prism at an angle θ_s, relative to the normal, given by $\sin\theta_s = k_s'(k_s'^2 + k_{sz}'^2)^{1/2}$.

The integral of Eq. (10) can be carried out analytically when $F(x)$ has a Gaussian form, i.e., $\boldsymbol{F}(x) = E_0 \exp(-4x^2/w^2)$. In this situation we obtain the following expressions for the intensity of the "reflected radiation"[9] in the prism and for $|E_x^t(\theta_i, x, z=-d)|$:

$$I^r(\theta_i, x, z=0) = |AF(x)\exp(ik_x^0 x) + iBG\exp(i\tilde{k}_s x)|^2, \tag{11}$$

$$|E_x^t(\theta_i, x, z=d)| = |\cos\theta_i CG\exp(i\tilde{k}_s x)|, \tag{12}$$

$$G = (w/2)\exp[-(k_x^0 - \tilde{k}_s)^2 w^2/16]\{1 + \mathrm{erf}[2x/w - i(k_x^0 - \tilde{k}_s)w/4]\}.$$

We note that for weakly damped SEM waves, i.e., $k_s'' \ll k_s'$, the dependence of G and, therefore, of $E_x^t(\theta_i, x, z=-d)$ on θ_i will exhibit sharp peaks at $k_x^0 = k_s'$ (i.e., at $\theta_i = \theta_s$), for values of x both within and beyond the excitation region. The reflected intensity $I^r(\theta_i, x, z=0)$, on the other hand, will exhibit ATR-type minima at $k_x^0 = k_s'$ for values of x within the excitation region where the radiative term is smaller in magnitude than the mirror reflection term, and will exhibit peaks for values of x where the radiative term is the dominant one. Furthermore, beyond the excitation region the magnitudes of the peaks in $I^r(\theta_i, x)$ will decrease exponentially with increasing x.

Calculations of the dependence of $E_x^t(\theta_i,x,z=-d)$ on x at $\theta_i=\theta_s$ for different values of w shows as expected that $|E_x^t/E_0|$ builds up to a maximum near the leading edge of the incident beam and decays exponentially beyond the excitation region. As w/L increases, the height of the maximum increases and approaches the value which is obtained for incident plane waves.

Finite beam width experiments were carried out with a 3-mm slit placed in the TM-polarized output beam of a cw CO_2 laser ($\lambda = 10.6$ μm) serving as the incident beam in a NaCl-air-Ag ATR configuration and with a second much narrower (0.125 mm) slit placed in front of a pyroelectric detector which could be translated in a direction normal to the mirror reflection axis. Measurements of the profile of the incident beam indicated that the beam profile was approximately Gaussian.

The experimental curve of the (normalized) power $P^r(\theta_i,x,w_c)$ versus x for $d = 38 \pm 2$ μm at $\theta_i=\theta_s=42°$ [where $P^r(\theta_i,x,w_c)=\int_{x-w_c/2}^{x+w_c/2} I(\theta_i,x')\,dx'$, and w_c is the lateral width of the detector slit] together with the corresponding theoretical curve based on $\epsilon_{\mathrm{NaCl}}=2.22$ and $\epsilon_{\mathrm{Ag}}=3850+i1450$ are given in Fig. 1(a). Apart from the magnitude of the minimum the experimental and theoretical curves are in relatively good agreement.[10] The double-peak structure is clearly evident, as is the exponential decay of P^r beyond the region of excitation. From the latter we obtain a value of 1.6 ± 0.05 mm for L, which is in reasonable agreement with the computer-calculated theoretical value of 1.66 mm for the leaky SEM wave.

Experimental curves of P^r versus θ_i for different values of x are given in Fig. 1(b). These show, as expected, sharp minima for values of x in the vicinity of $x=0$ and sharp peaks for values of x beyond the excitation region, whose magnitudes decrease with increasing x.[11]

It is the occurrence of peaks in P^r versus θ_i curves for x beyond the excitation region, signaling the presence of free leaky SEM waves, that is the most significant feature of the excitation of SEM waves by VEM waves of finite beam width. We note furthermore that it is the leading edge of the incident VEM radiation that is responsible for the occurrence of the reflected minima and the exponential decay of the reflected intensity. The presence of the back edge primarily determines the amplitude of the excited SEM waves.[12]

In conclusion, we note that the excitation of SEM waves by finite beams in single-prism configuration can be used to determine the propagation wave vector (i.e., k_s') and the propagation length of SEM waves. Moreover, the method is a versatile one, since such data can be obtained using both the Kretschmann and the Otto prism

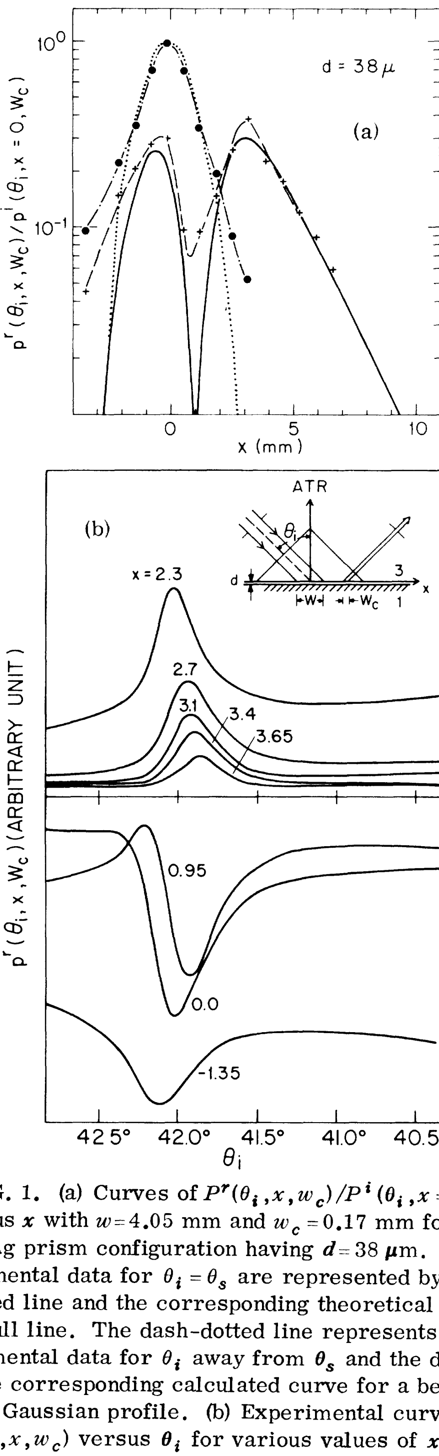

FIG. 1. (a) Curves of $P^r(\theta_i,x,w_c)/P^i(\theta_i,x=0,w_c)$ versus x with $w=4.05$ mm and $w_c=0.17$ mm for a NaCl-air-Ag prism configuration having $d=38$ μm. The experimental data for $\theta_i=\theta_s$ are represented by the dashed line and the corresponding theoretical curve by the full line. The dash-dotted line represents the experimental data for θ_i away from θ_s and the dotted line is the corresponding calculated curve for a beam having a Gaussian profile. (b) Experimental curves of $P^r(\theta_i,x,w_c)$ versus θ_i for various values of x in millimeters. The data are obtained with a 3-mm slit in the incident beam and a 0.3-mm slit in front of the detector.

configurations.

We wish to acknowledge valuable discussions with Y. J. Chen and A. Hjortsberg.

*Research supported by the U. S. Office of Naval Research and the National Science Foundation.

[1]E. Burstein, W. P. Chen, Y. J. Chen, and A. Hartstein, J. Vac. Sci. Technol. 11, 1004 (1974), and references therein.

[2]C. A. Ward, R. W. Alexander, and R. J. Bell, Phys. Rev. B 12, 3293 (1975).

[3]Y. J. Chen, W. P. Chen, and E. Burstein, Phys. Rev. Lett. 36, 1207 (1976).

[4]A. Otto, Z. Phys. 216, 398 (1968).

[5]E. Kretschmann, Z. Phys. 241, 313 (1971).

[6]J. Schoenwald, E. Burstein, and M. Elson, Solid State Commun. 12, 185 (1973).

[7]J. D. McMullen, Solid State Commun. 17, 331 (1975).

[8]D. L. Begley, D. A. Bryan, R. W. Alexander, R. J. Bell, and C. A. Goben, to be published.

[9]T. Tamir and H. L. Bertoni, J. Opt. Soc. Am. 61, 1397 (1971).

[10]The discrepancy between the theoretical and experimental curves is due to roughness scattering and surface-contamination effects and to the deviation of the incident beam from a Gaussian profile.

[11]We have also observed a similar excitation of leaky modes at $\theta_i < \theta_{\rm critical}$ corresponding to "virtual" modes within the air gap between the prism and metal which have $\tilde{k}_{2z}$ predominantly real.

[12]It is also obvious that similar effects will occur with excitation of SEM waves by finite-beam-width VEM waves in grating-coupling configurations.

"Grooving" of a Uniform Plasma with a Magnetic Field by an Unstable Wave

Tetsuya Sato
Geophysics Research Laboratory, University of Tokyo, Tokyo 113, Japan
(Received 4 June 1976)

Presented numerically is an apparently paradoxical nonlinear process that a two-stream instability is stabilized by inhomogenizing or "grooving" an initially homogeneous plasma, not by limiting the stream which is the direct cause of the instability. For a relatively wide and slow electron stream, the instability is shut off by reducing the speed to the threshold value (ion sound speed), as is naturally expected. For a narrow and fast stream, however, the instability is shown to be stabilized by the grooving effect.

From the standpoint of causality, one may say that an instability proceeds in general so as to relax its driving force. For instance, drift waves would be stabilized by flattening the density gradient which causes the instability. Recently, however, an apparently paradoxical stabilization mechanism has been proposed for a resistive two-stream (Hall current) instability.[1] This instability is widely known among ionospheric plasma physicists as the two-stream instability or "type I" irregularities.[2] The only driving force of this instability in a uniform plasma is the electron stream. It may therefore be natural that one considers the instability to be stabilized by self-limitation of the stream. In fact, it has been shown that an unstable wave itself can reduce the electron drift speed to the threshold (ion acoustic) speed.[3] The recent theory,[1] however, indicates that the two-stream instability can also be stabilized by "inhomogenizing" a uniform plasma. If this process, which does not take part in the linear instability mechanism, is indeed capable of stabilizing the two-stream instability, then one must recognize a strange fact that the nonlinear development of an instability is not necessarily connected by one causal chain but may happen to be governed by an apparently irrelevant causality.

Let us consider a magnetized, uniform plasma in which a Hall current (primarily carried by Hall electrons) is concentrated in a limited layer. I use a coordinate system (x, y, z), the z axis being directed antiparallel to the magnetic field $\vec{B}_0$, the y axis being directed parallel to the electron drift, and the x axis being directed so as to form a right-handed system.

I assume slightly collisional electrons ($\nu_e/\Omega_e \ll 1$) and heavily collisional ions ($\nu_i/\Omega_i \gg 1$), ν_e and ν_i being the collision frequencies of electrons and ions (against neutrals), respectively, and Ω_e Ω_i being the gyrofrequencies of electrons and ions, respectively. Accordingly, the motion of electrons is dominated by the electric (Hall) drift, while the motion of ions is almost completely controlled by collisions (hence no appreciable ion Hall drift is present). Furthermore, I assume a low-frequency electrostatic mode which propa-