

regime determines the stability of the system. This is not surprising since it is this regime which is unstable in the case of a static plasma. Thus we next consider $kR \ll \delta$ and expand Δ first in k and then in δ . The leading term is of order $k^0 \delta^2$ and is given by

$$\Delta_{0,2} = AT \int_0^\lambda \left(\frac{dR}{dz} \right)^2 dz,$$

where A is a positive constant and

$$T = 4 \left(\frac{V}{B} \right)^4 - \left\{ (m-1) + (m-3) \left(\frac{V}{B} \right)^2 \right\} \frac{\left\{ (m-1) + (m-3) (V/B)^2 \right\}^2}{1 - (V/B)^2},$$

where V and B are zero-order quantities in δ and therefore constants.

T may also be written in the form

$$T = \frac{1}{4} \left\{ \left[4 \left(\frac{V}{B} \right)^2 - \frac{1}{8} \right]^2 + \frac{63}{64} \right\} - \frac{1}{1 - (V/B)^2} \times \left\{ (m-1) + (m-3) \left(\frac{V}{B} \right)^2 + \frac{1}{2} \left[1 - \left(\frac{V}{B} \right)^2 \right] \right\}^2.$$

It is clear that if $V^2 > B^2$, then $T > 0$, $\Delta_{0,2} > 0$, and therefore $\Delta > 0$ to all orders in δ and ϵ .

This result therefore shows that for $\delta \gg \epsilon$, the plasma is stable to all $m \geq 1$ provided that $V^2 > B^2$. In physical terms this means that a static plasma with a periodic profile, for which all modes $m > 1$ would be unstable, will be stable for all $m \geq 1$ if the profile is made to prop-

agate along the plasma as a wave with velocity greater than the "Alfvén speed."

The implication of this for the toroidal theta pinch is that one is led to conjecture that the equilibrium profile, which is necessarily periodic and therefore unstable, may be stabilized if the profile is made to propagate around the torus with sufficiently high velocity.

It should be noted that the $m = 0$ mode is not covered by the present analysis and although this mode is stable without dynamic stabilization, the possibility of introducing instability cannot be ruled out.

It is also possible that although the present model for the plasma allows the wave to propagate without accelerating the plasma, this may not be the case for an actual plasma. This leads to the question of whether similar stabilization can be achieved by a standing wave rather than a propagating wave. Consideration is now being given to this problem.

¹J. A. Wesson, in Proceedings of the Conference on Pulsed High Density Plasmas, Los Alamos, 1967 (to be published).

²F. Meyer and H. U. Schmidt, Z. Naturforsch. **13a**, 1005 (1958).

³F. A. Haas and J. A. Wesson, to be published.

⁴F. A. Haas and J. A. Wesson, Phys. Fluids **9**, (1966).

⁵E. S. Weibel, Phys. Fluids **3**, 946 (1960).

⁶E. Frieman and M. Rotenberg, Rev. Mod. Phys. **32**, 898 (1960).

TOTAL REFLECTION IN SECOND-HARMONIC GENERATION*

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We have observed the reflected second-harmonic intensity generated by an incident beam which is totally reflected by the nonlinear medium immersed in a denser linear medium. The reflected intensity becomes very high near the critical angle for total reflection. The role of momentum matching for the reflected intensity is demonstrated.

The theory of the reflected harmonic intensity¹ has been verified experimentally^{2,3} for a variety of geometrical configurations. The purpose of this note is to present experimental data for the interesting geometry that the fundamental beam is incident from a dense linear medium and is totally reflected by the nonlinear medium. Singularities predicted by the theory¹ are demonstrated experimentally and anomalously high reflected harmonic in-

intensities are obtained because phase matching is important for the reflected intensity at the critical angle for total reflection.

The stimulated Stokes beam induced by a Q-switched ruby laser in H₂ gas is passed through a fluid cell containing 1-bromonaphthalene. This optically dense fluid has an index of refraction $n(\omega) = 1.628$ and $n(2\omega) = 1.682$, where the second-harmonic frequency 2ω corresponds to a wavelength of 4860 Å. The second harmon-

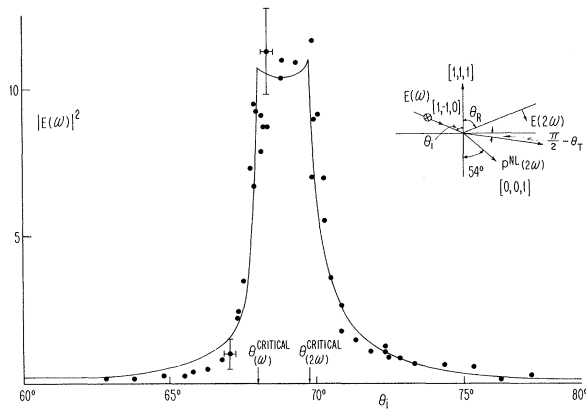


FIG. 1. The reflected harmonic intensity from NaClO₃ in the neighborhood of the critical angle for total reflection. The incident beam at λ = 9720 Å passes through the optically dense fluid 1-bromonaphthalene. The angle of incidence θ_i is varied in the geometry shown in the inset. The drawn curve is calculated theoretically and has cusps at the critical angles for total reflection.

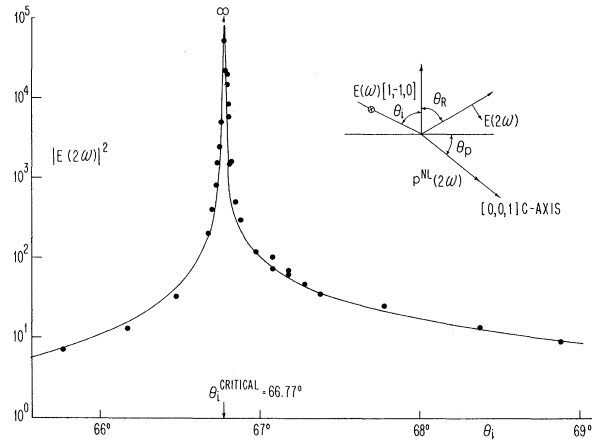


FIG. 2. The reflected harmonic intensity from KH₂PO₄ in the neighborhood of the critical angle for total reflection. The phase-matched direction is parallel to the surface. The incident beam at λ = 9720 Å passes through the optically dense fluid 1-bromonaphthalene. The angle of incidence θ_i is varied in the geometry shown in the inset. The drawn curve is calculated theoretically (see text).

ic of ruby light itself, at 3470 Å, is absorbed by this fluid. In this fluid a nonlinear crystal of NaClO₃ or KH₂PO₄ is immersed.

The reflected second-harmonic intensity is observed as a function of the angle of incidence θ_i for the crystallographic orientation and polarization directions shown in the inserts of Figs. 1 and 2. The incident beam had a rectangular cross section of 2 × 3 mm and had an angular divergence less than 2 mrad. The power density was about 5 MW/cm². The front

face of the crystals was flat to ½λ over 1 cm and the back face was ground to avoid spurious reflections.

The theory for second-harmonic generation in a piezoelectric crystal of cubic symmetry gives the following expression for the reflected intensity polarized in the plane of reflection¹:

$$g(2\omega) = 4\pi |\chi_{14}^{NL}|^2 |E_{inc}(\omega)|^2 |F^{NL}|^2 |F^L|^4. \quad (1)$$

Here the nonlinear Fresnel factor is given by

$$F^{NL} = \frac{\sin\theta_S \sin^2\theta_T \sin(\alpha + \theta_S + \theta_T)}{\epsilon_R(2\omega) \sin\theta_R \sin(\theta_T + \theta_S) \sin(\theta_T + \theta_R) \cos(\theta_T - \theta_R)} \quad (2)$$

and the linear Fresnel factor for transmission is

$$F^L = \frac{2 \sin\theta_S \cos\theta_i}{\sin(\theta_i + \theta_S)}. \quad (3)$$

The angles θ_R, θ_S, and θ_T are related to θ_i by

$$\begin{aligned} \sin\theta_R &= [n_{liq}(\omega)/n_{liq}(2\omega)] \sin\theta_i, \\ \sin\theta_S &= [n_{liq}(\omega)/n_{cr}(\omega)] \sin\theta_i, \\ \sin\theta_T &= [n_{liq}(\omega)/n_{cr}(2\omega)] \sin\theta_i, \end{aligned} \quad (4)$$

where n_{cr} stands for refractive index of either

KH₂PO₄ or NaClO₃ and n_{liq} for that of 1-bromonaphthalene. α + θ_S is the angle between the nonlinear polarization and the normal to the surface, which is constant in the geometries used.

The index of refraction for NaClO₃ at the wavelengths of interest is n(ω) = 1.509 and n(2ω) = 1.528. The small effect of natural optical activity may be ignored in this experiment. The nonlinear susceptibility⁴ is χ₁₄(NaClO₃) = 1.7 χ₁₄(KH₂PO₄).

The drawn theoretical curve, calculated from Eqs. (1) to (4), is compared with the experimental results in Fig. 1. Note the striking agree-

ment with the theoretical curve, which displays nonanalytical singularities at angles of $\theta_i = 67.96^\circ$, for which $\sin\theta_S = 1$, and $\theta_i = 69.81^\circ$, for which $\sin\theta_T = 1$. At these angles, $\cos\theta_S$ and $\cos\theta_T$, respectively, change from a real value to a pure imaginary value.

The behavior of the reflected intensity in the neighborhood of these critical points is dominated by the term

$$|\sin(\theta_T + \theta_S)|^{-2} \\ = |\sin\theta_T(1 - \sin^2\theta_S)^{1/2} + \sin\theta_S(1 - \sin^2\theta_T)^{1/2}|^{-2}.$$

For $\sin\theta_T = 1$ the magnitude of this factor is $n_{cr}^2(\omega)/\{n_{cr}^2(\omega) - n_{cr}^2(2\omega)\}$. Near the critical angle of total reflection the reflected harmonic intensity is larger than the intensity away from this angle by a factor $\epsilon_S(\epsilon_T - \epsilon_S)^{-1}$, and has an order of magnitude equal to the geometric mean of the normal reflected intensity and the maximum intensity generated in transmission through a plane parallel platelet.⁵ The maximum reflected intensity in NaClO₃ is about 2% of this maximum transmitted intensity, in good agreement with the above theoretical considerations.

The physical interpretation is that momentum matching^{6,7} for the wave propagating parallel to the surface inside the nonlinear medium is important in determining the reflected intensity at the critical angle. The momentum mismatch occurs in the direction normal to the surface and the reflected second harmonic near the critical angle is generated by the polarization in a surface layer with a thickness of about $(\lambda l_{coh})^{1/2}$.

These results suggest that in a uniaxial crystal where a direction of perfect phase matching is parallel to the surface, the reflected intensity should tend to infinity when the angle for total reflection is approached. A KH₂PO₄ crystal was cut with the *c* axis making an angle $\theta_p = 42^\circ 14'$ (phase-match angle for the wavelengths used) with the surface. The experimental results in Fig. 2 show that the second-harmonic reflected intensity increases by about

three orders of magnitude when the angle of incidence is changed by a few tenths of a degree. The observed maximum reflected intensity is two orders of magnitude larger than the maximum transmitted intensity in NaClO₃. The observed maximum value is determined by the width *d* or diffraction aperture of the incident beam. When $l_{coh} = \infty$, the maximum depth of the contributing nonlinear layer is $(\lambda d)^{1/2}$. The intensity of the reflected second harmonic then goes as d^2 , whereas normally it is, of course, just proportional to the cross section of the beam, i.e., proportional to *d*. This has been verified experimentally.

The drawn curve in Fig. 2 has been calculated using again Eqs. (1)-(4) valid for a cubic crystal with $\theta_S = \theta_T$ and assuming $n(\omega) = n(2\omega) = 1.496$ without including angular variations. This is not rigorously correct for a uniaxial crystal. Complete, but necessarily very involved, expressions have been given by Fisher.⁸ It is seen that the variation in the immediate neighborhood of the critical angle is rather well described by Eqs. (1)-(4) for the geometry used. A more detailed analysis of this case will be given elsewhere.⁹ The dominant factor is again $|\sin(\theta_S + \theta_T)|^{-2}$ which tends to approach infinity for $\theta_S = \theta_T = \frac{1}{2}\pi$.

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¹N. Bloembergen and P. S. Pershan, Phys. Rev. **128**, 606 (1962).

²R. K. Chang and N. Bloembergen, Phys. Rev. **144**, 775 (1966).

³N. Bloembergen, Opt. Acta **13**, 311 (1966), and references quoted therein.

⁴N. Bloembergen and J. Simon, Bull. Am. Phys. Soc. **12**, 687 (1967).

⁵N. Bloembergen, *Nonlinear Optics* (W. A. Benjamin, Inc., New York, 1965), p. 82.

⁶J. A. Giordmaine, Phys. Rev. Letters **8**, 19 (1962).

⁷P. D. Maker, R. W. Terhune, and M. Nisenoff, Phys. Rev. Letters **8**, 21 (1962).

⁸R. Fisher, Phys. Status Solidi **19**, 757 (1967).

⁹C. H. Lee, thesis, Harvard University, 1967 (unpublished).