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[2]See, for example, A. Abragam, The Principles of Nuclear Magnetism (Clarendon Press, Oxford, England, 1961).
[3]R. W. G. Wyckoff, Crystal Structures (Interscience Publishers, Inc., New York, 1964), Vol. 2.
[4]In fact, the line begins to broaden again at higher temperatures. This effect was first pointed out by M. Goldman and L. Shen who will publish more detailed high-temperature data and an explanation of this effect.
[5]These measurements will be reported in detail elsewhere.
[6]D. A. Jones, J. M. Baker, and D. F. D. Pope, Proc. Roy. Soc. (London) 74, 249 (1959); R. A. Buchanan and H. H. Caspers, Naval Weapons Evaluation Facility Report No. 8213, 1964 (unpublished); W. M. Yen, W. C. Scott, and A. L. Schalow, Phys. Rev. 136, 271 (1964).

NONLINEAR OPTICAL REFLECTION FROM A METALLIC BOUNDARY*

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Of the many nonlinear optical effects reported since Franken's discovery,[1] none has confirmed the generation of optical harmonics by a conduction electron plasma. Although Ducuing and Bloembergen[2] have studied harmonics generated by semiconducting mirrors, the observed effects are not related to the motion of conduction electrons, occurring only for noncentrosymmetric crystals and for power levels several orders of magnitude less than those here employed. The purpose of the present Letter is to report the unambiguous observation of second-harmonic light generated on reflection of a giant-pulse laser beam from the surface of a silver mirror. The present situation is distinct from earlier cases of optical harmonic generation, because the host lattice is centrosymmetric and the lack of symmetry of the reflecting boundary is responsible for the harmonic production. The observations are consistent with a second-harmonic polarization proportional to $\vec{E}\nabla\vec{E}$. Since only the component of $\vec{E}$ normal to the surface has a discontinuity at the boundary, the second-harmonic polarization would be proportional to $\cos^2\theta$, where θ is the angle between $\vec{E}$ and the plane of incidence. The second-harmonic intensity would be proportional to $\cos^4\theta$.

Figure 1 describes the apparatus used to detect the effect and measure its angular dependence. The principal feature is the prism assembly consisting of three right-angle prisms with evaporated silver mirror surfaces mounted together as a unit. A ray incident on one side of the assembly then undergoes specular reflection at four successive surfaces and emerges in a direction colinear with the original beam. The arrangement permits both laser and monochromator to remain fixed, while θ can be varied by rotating the prism assembly around the beam as axis. The filter inserted after the third reflection eliminates harmonic light from all but the last surface and limits observed effects to those associated with reflection from a sin-

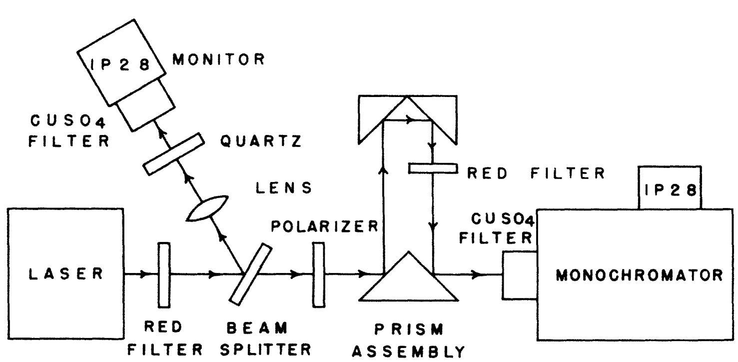

FIG. 1. Diagram of apparatus.

gle mirror. For θ other than 0° or 90°, the incoming beam is then composed of two components parallel and perpendicular to the plane of incidence. Each of these traverses the assembly independently of the other and emerges displaced in relative phase due to the several metallic reflections. In the above model, however, the component perpendicular to the plane of incidence (which plane is the same for all surfaces) produces no second harmonic and can be ignored regardless of its phase with respect to the other component lying in the plane of incidence. The latter is assumed to be the sole driving source effective in producing harmonics. The intensity of the harmonic light from the last mirror in the assembly therefore varies with θ according to $\cos^4\theta$, as predicted for simple reflection from a single surface. The ruby laser itself produces pulses of the order of 1 MW peak power and <50 nsec duration by means of a saturable-dye Q spoiler (cryptocyanine).[3] The power is monitored by dual-beam oscilloscope comparison of the signal with an auxiliary second-harmonic pulse produced in quartz.[2]

Second-harmonic signals seen with the above apparatus consist of sharp, clean pulses similar in length to those of the monitor and several orders of magnitude above the limiting noise of the system. In order to avoid conflicting background radiation, it has been found necessary to use evaporated silver layers thick enough to be thoroughly opaque. Laser power must likewise be restricted to the 1-MW level in order to prevent film destruction and concurrent thermal radiation. With these precautions the harmonic signal passes the usual tests: (1) It disappears when the sample (prism assembly) is removed, the primary beam being undeviated; (2) it occurs uniquely at 3473 ± 10 Å; (3) the signal is coincident (to $\approx$10 nsec) with the monitor and has the same pulse shape. Occasionally, for silver films too thin or for excessive input power, signal pulses are longer than the monitor pulses and take on a characteristic irregular appearance. The true signal is then submerged by background radiation over a broad band of wavelengths. Harmonic generation efficiency is about 10^{-15}.

The symmetry of the effect is clearly demonstrated by the data of Fig. 2, which indicates the pronounced dependence of harmonic intensity on the normal component of $\vec{E}$. The $\cos^4\theta$ curve, expected theoretically for the above model, is included in the figure for comparison. Agreement is satisfactory.

Since background effects are troublesome at these power levels, it is important to establish that the present observations are not due to adsorbed molecules on the silver surface or due to other surface contamination. It is well known[4] that silver does not adsorb N_2, CO, CO_2, Ar, or H_2 under ordinary laboratory conditions. Studies have shown[5,6] that the adsorption of oxygen by silver catalysts or by evaporated silver is thermally activated and does not occur to a measurable extent under the conditions of the present experiment, namely, 25°C and atmospheric pressure. Adsorption of oxygen does occur, however, above about 180°C, although even at this temperature the reaction is slow and reaches a saturation value of 0.6 to 0.9 monolayers on the surface only after 20 minutes or more. As a double precaution against adsorption, we have tested silver layers evaporated at 10^{-5} Torr and kept subsequently under argon. Films were transferred from the evaporator to the argon-filled test chamber in less than one minute. Under these circumstances observed second-harmonic signals are substantially the same as those found for films exposed to air for one hour after preparation, and also for those exposed for as much as three weeks. In another precautionary experiment, we have looked for harmonic light from fused quartz plates which had been exposed to the vacuum-chamber atmosphere at 10^{-5} Torr, where thin contaminating layers are known to be deposited.[7] Since no such light has been seen, we believe that contamination does not contribute to the harmonic signals reported here. It is therefore concluded that the present radiation comes from the silver itself and is not produced artificially by a layer adsorbed physically or chemically on the surface.

It is important to distinguish the present situation from the one discussed by Bloembergen.[8] In the latter case the plasma vibrates at 2ω in the direction of the incident beam, and any observed harmonic radiation must be approximately independent of incident-beam polarization. In view of the strong polarization dependence demonstrated in Fig. 2, it is clear that light emitted by such a mechanism cannot contribute more than about a tenth of the presently observed signal. Higher order nonlinear multipole radiation has been assumed negligible.

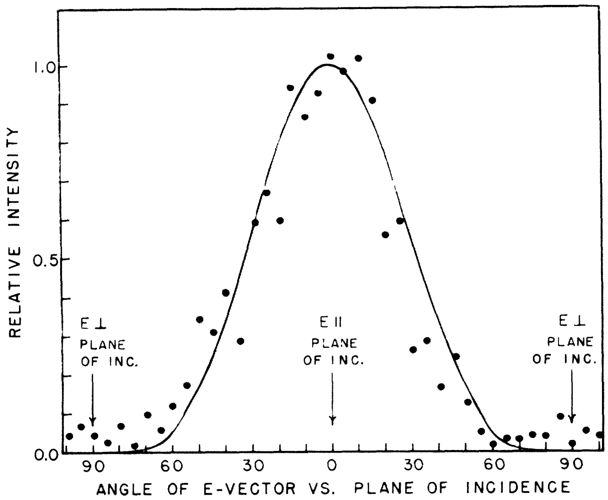

FIG. 2. Variation of second-harmonic light intensity with angle θ between the incident-light polarization vector and the plane of incidence. The theoretical $\cos^4\theta$ dependence (solid line) is included for comparison.

The comparison between the nonlinear susceptibility of conduction electrons and that of bound electrons in crystals such as quartz or potassium dihydrogen phosphate is clear. In both cases second-harmonic effects are enhanced when the system resonates at a frequency near 2ω. In the case of silver irradiated with ruby light, the plasma resonance is close to 2ω, making it a favorable material for initial experimentation. It will be of interest to study second-harmonic light produced in a variety of metals under uniform surface conditions in order to test the applicability of resonance theory to the case of harmonic generation by plasma oscillations.

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PION-PROTON TOTAL CROSS SECTIONS: 400 TO 2000 MeV

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We have measured the total cross section for the scattering of positive and negative pions on protons in the energy range of 400 MeV to 2 GeV with a statistical uncertainty of about 1% (0.3 to 0.5 mb). Known systematic uncertainties are of the same order or less except at energies below 750 MeV for π^--p where they vary from 2 to 10%.

The primary aim of this work was to resolve the systematic 3- to 4-mb difference between the Saclay[1] and Berkeley[2] measurements of the π^+-p total cross sections over most of this energy range. There was also some need to resolve differences in the measurements of the π^--p cross sections, especially at the peaks and valleys in the vicinity of the second and third "resonances." We also sought detailed measurements in the vicinity of the 830-MeV shoulder in the π^+-p cross section, and in the region of 1.5 to 2.0 GeV where a comparison with the Brookhaven measurements[3] was possible.