pother, and M. Rayl, Phys. Rev. Letters 19, 357 (1967).
[11]We consider here the worst case: $p \equiv 1$. The exponent η is expected to be small; see Refs. 3 and 8, and M. E. Fisher and R. J. Burford, Phys. Rev. 156, 583 (1967).
[12]Fisher and Burford, Ref. 11.
[13]D. Bally, B. Grabcev, A. M. Lungu, M. Popovici, and M. Totia, J. Phys. Chem. Solids 28, 1947 (1967).
[14]This singularity is not specifically shown in Figs. 10 and 12 of Ref. 11 but its presence follows from the considerations presented here.
[15]M. E. Fisher, in Critical Phenomena, Proceedings of a Conference, Washington, D. C., 1965, edited by M. S. Green and J. V. Sengers, National Bureau of Standards Miscellaneous Publication No. 273 (U. S. Government Printing Office, Washington, D. C., 1966), pp. 108-115, especially Eqs. (40)-(45).
[16]For small $k_F l$ the opposite sign for the resistive anomaly cannot be excluded. It is also possible that the approximation of the sum in (13) by an integral might be misleading in this and certain other circumstances.
[17]B. R. Coles, Advan. Phys. 7, 40 (1958).
[18]R. V. Colvin, S. Levgold, and F. H. Spedding, Phys. Rev. 120, 741 (1960); H. E. Nigh, S. Levgold, and F. H. Spedding, Phys. Rev. 132, 1092 (1963).

SECOND-HARMONIC GENERATION OF LIGHT AT THE BOUNDARY OF ALKALI HALIDES AND GLASSES

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This Letter reports measurements of second-harmonic generation of light at the boundary of alkali halides and glasses along with further measurements of some semiconductors and metals. The second-harmonic power reflected from the surface of transparent materials was found to be lower by several orders of magnitude than that generated at semiconductor and metallic surfaces.[1-4] Second-harmonic generation was also studied in transmission through thin platelets of glass and LiF.

The second-harmonic generation in media with inversion symmetry has been discussed by several authors.[5-10] For centrosymmetric isotropic medium, the lowest order contribution to the nonlinear polarization at the second-harmonic frequency 2ω, denoted by $P_i(2\omega)$, may be given phenomenologically as

$$P_i(2\omega) = (\alpha-\beta)E_j(\omega)\nabla_j E_i(\omega) + \beta E_i(\omega)\nabla_j E_j(\omega) + \gamma\nabla_i[E_j(\omega)E_j(\omega)]. \quad (1)$$

The coefficients are expected to be of the order of 10^{-17} esu for transparent materials,[5] about 10^{-15} esu in semiconductors, and 10^{-14} esu in metals.[9] All materials will be assumed to be isotropic, as no signals due to the additional terms present with cubic crystals have been observed in this or previous experiments.[1]

For a transverse electromagnetic wave, the first two terms in Eq. (1) are zero in the bulk; the last term gives rise to a longitudinal polarization, and so it is observable only in experiments involving a discontinuity, such as the boundary of a medium. This bulk term alone cannot explain the observed effects, and a proper account must include phenomena associated with the surface itself. To this end, the model of Bloembergen[2,10] will be used. According to this model, the nonlinear surface effects arise primarily from the rapid variation of the normal component of the electric field over a thin layer at the boundary of the medium. Assuming that the thin layer has the same dielectric constant as that of the bulk, this model predicts that the surface effects can be described in terms of a nonlinear polarization equivalent to the α and β terms in Eq. (1). For layers thin compared with a wavelength, the corresponding second-harmonic amplitude depends on the total change of the normal component across the layer and is independent of the detailed variation within the layer.

An alternative model to describe the surface effects is to assume that the surface is piezoelectric.[1] General symmetry considerations show that the only new term distinguishable from the previous model is of the form $P_{\text{normal}}(2\omega) \propto E_j E_j$. A slightly different angular dependence not detectable in this experiment would follow from a term of this form.

The coefficients α, β, and γ are generally complex, but are real in a lossless medium. These coefficients can be determined by measuring the power and polarization of the second harmonic produced with the fundamental beam polarized at 45° and 90° with respect to the plane of incidence. Using Eq. (1) and as-

suming the existence of a strong gradient of the normal component of the electric field in a thin layer at the boundary,[10] one calculates the second-harmonic power reflected from an air-dielectric interface[11] as follows:

$$P_{\perp}^{1/2} = (16\pi^2/\lambda)\cos^2\theta_i |\beta(\epsilon_S - 1)\sin\theta_S/(g_{1S}g_{0S}g_{1T})|E_0{}^2A^{1/2}, \tag{2}$$

$$\left(\frac{P_\gamma}{P_\perp}\right)^{1/2} = \left|\frac{\gamma}{\beta(\epsilon_S - 1)}\left(\frac{\epsilon_S}{\epsilon_T}\right)^{1/2}\frac{g_{0S}g_{1T}}{g_{1S}g_{0T}}\right|, \tag{3}$$

$$\left(\frac{P_\parallel}{P_\perp}\right)^{1/2} = \left|\frac{g_{1S}g_{1T}}{g_{0S}g_{0T}}\right|\left|\left[\left(1+\frac{g_{0S}{}^2}{g_{1S}{}^2}\right)\left(\frac{\gamma}{\beta}\right)\frac{\epsilon_S}{\epsilon_S - 1} - \frac{\alpha}{\beta}\sin^2\theta_i\right](\epsilon_S\epsilon_T)^{-1/2} + \cos\theta_S\cos\theta_T\right|. \tag{4}$$

Here the fundamental beam has an area A, an electric field amplitude E_0, and an angle of incidence θ_i. The wavelength of the second-harmonic radiation is denoted by λ. The components of the second-harmonic power polarized parallel and perpendicular to the plane of reflection corresponding to the fundamental beam polarized at 45° to the plane of incidence are $P_\parallel$ and $P_\perp$, respectively. The second-harmonic power generated with the fundamental beam polarized perpendicular to the plane of incidence and always polarized in the plane of reflection is denoted by P_γ. The angle of refraction is denoted by θ; the dielectric constant by ϵ; the subscripts S and T refer, respectively, to the fundamental and second-harmonic frequencies; $g_0 = \epsilon^{1/2}\cos\theta_i + \cos\theta$; and $g_1 = \epsilon^{1/2}\cos\theta + \cos\theta_i$.

The experiments were performed with a ruby laser Q switched by a rotating prism and fired at a repetition rate of one pulse per second. The output from the laser was approximately 30 mJ in energy, 0.4×10^{-2} cm^2 in cross section, and 100 nsec in duration. For reflection measurements, the samples were placed in a convergent beam half-way between a 50-cm focal length lens and its focus. For KI and other high-index materials, it was necessary to attenuate the laser beam to prevent dielectric breakdown either at the sample surfaces or within the samples. Individual photoelectrons were detected using a high-gain photomultiplier and an analog sampling circuit.[12] Typical signal level for transparent materials amounted to about 20 photoelectrons per 1000 laser shots, with the background count less than 4 photoelectrons per 1000 shots. The measurements of harmonic power to be reported herein represent an average over as many as up to 5000 laser shots; so the uncertainty for each measurement was reduced to 10% or less.

For alkali-halide crystals, no difference was observed between results obtained with polished (100) surfaces and with cleaved surfaces. The semiconductor surfaces were lapped and etched to minimize work damage at the surface.[13] Samples of single-crystal silver and gold were electropolished in a mixture of 95% ethanol and 5% phosphoric acid and in KCN solution, respectively. The results were reproducible to within about 15% for all materials independently of the surface treatments.

The measurements of reflected second-harmonic power are summarized in Table I. With the fundamental beam polarized perpendicular to the plane of incidence, the second-harmon-

Table I. Second-harmonic power generated at the boundary of centrosymmetric isotropic media and cubic crystals. The angle of incidence for the fundamental beam was 60°.

Material	Second-harmonic conversion efficiency (10^{17}) $P_\perp$	P_γ	$P_\parallel/P_\perp$
CaF_2	5.5	a	0.9
BaF_2	6	a	0.8
NaF	8.5	a	0.96
LiF	8	a	0.8
Fused silica	8.5	a	0.96
BSC glass	8	a	0.9
KBr	6.5	a	0.73
KCl	12	a	0.61
NaCl	9.5	a	0.4
KI	25	1.4	0.31
MgO	35	4	0.11
Si	110	120	5.1
Ge	60	160	17
Au	320	240	11
Ag	3000	1900	13

[a]Estimated to be 1.0 or less.

ic wave was found to be polarized in the plane of reflection for all materials. With the fundamental beam polarized at 45° with respect to the plane of incidence, the harmonic wave for transparent materials was linearly polarized with the E vector lying in the same quadrant as the fundamental beam, but for metals and semiconductors, it was slightly elliptically polarized. In the latter cases, no effort was made to determine accurately the ellipticity and orientation of the polarization. Using the refractive-index data available in the literature,[14] the values of α, β, and γ deduced according to Eqs. (2)-(4) are listed in Table II. The data were insufficient for deducing α/β in metals and semiconductors.

Second-harmonic generation was also studied in transmission through thin platelets of glass and LiF. When the platelet was immersed in index-matching liquid, the second-harmonic power generated was reduced by about a factor of 400 from that generated when the platelet was in air. Figure 1 shows the second-harmonic power generated in transmission through a glass platelet in air as a function of the angle of incidence. The experimental points can be described fairly well by an equation of the following form:

$$\text{second-harmonic generation} \propto (2\cos\theta_i/g_{0S})^4 \sin^2(\Delta\varphi)\sin^2\theta_i \times[4\sin\theta_S\sin\theta_T+\cos\theta_S\cos\theta_T]^2/(\epsilon_T^{1/2}\cos\theta_i+\cos\theta_T)^2, \quad (5)$$

where $\Delta\varphi=(\omega d/c)(\epsilon_S^{1/2}\cos\theta_S-\epsilon_T^{1/2}\cos\theta_T)$ and d is the thickness of the platelet. Equation (5) may be obtained from the analysis of Bloembergen and Pershan[11] by neglecting the interference due to the channel spectra at the harmonic frequency and setting $\alpha/\beta=4$ and $\gamma=0$. The data represent poor averaging over the rapid modulation with angle of the generated harmonic power. This modulation arises from the multiple reflections of the waves in the platelet at both the fundamental and harmonic frequencies. These interference effects rendered it difficult to obtain quantitative information from the transmission measurements.

Table II. Values of the nonlinear susceptibility constants α, β, and γ deduced from the harmonic-power measurements listed in Table I.

Material	α/β	β (10^{16} esu)	γ (10^{16} esu)
CaF_2	4.0	0.4	a
BaF_2	4.0	0.4	a
NaF	3.4	0.5	a
LiF	3.6	0.5	a
Fused silica	4.2	0.5	a
BSC glass	4.5	0.5	a
KBr	4.6	0.45	a
KCl	4.0	0.6	a
NaCl	3.9	0.55	a
KI	4.5	1.0	0.2
MgO	4.2	1.2	0.25
Si	b	6.9	70
Ge	b	4.9	85
Au	b	3.0	15
Ag	b	5.3	20

[a]Estimated to be less than 0.1.
[b]See text.

The values of the coefficients in Table II are of the expected order of magnitude for all materials. It is interesting to observe that for transparent materials, the ratio α/β is found to be approximately 4 within experimental accuracy independently of the surface treatments. Also, the coefficients seem to depend monotonically upon the index discontinuity at the interface. This idea is supported by the observation that second-harmonic generation was

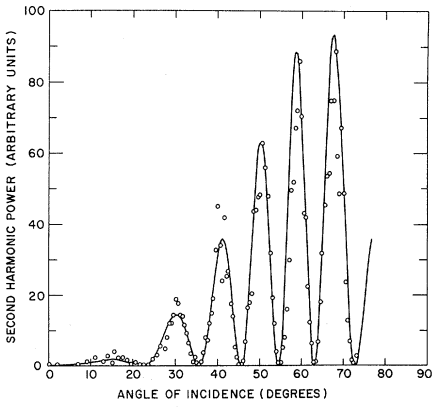

FIG. 1. Second-harmonic power generated in transmission through a glass platelet as a function of angle of incidence. The experimental points were obtained with an unfocused laser beam polarized in the plane of incidence. The solid curve is a plot of Eq. (5) with $\lambda=3472$ Å, $d=2.12\times10^{-2}$ cm, $n=1.5219$, and $\Delta n=0.031$.

not detectable under index-matching conditions. At present, theory remains to be developed to explain these various effects. The present experimental capability also needs to be refined to observe deviations of α/β from the constant value, thus providing a new tool for studying the optical properties of surfaces.

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[1]N. Bloembergen, R. K. Chang, and C. H. Lee, Phys. Rev. Letters 16, 986 (1966); C. H. Lee, R. K. Chang, and N. Bloembergen, Phys. Rev. Letters 18, 167 (1967).

[2]N. Bloembergen and R. K. Chang, in Physics of Quantum Electronics, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Company, Inc., New York, 1966), p. 80.

[3]F. Brown, R. E. Parks, and A. M. Sleeper, Phys. Rev. Letters 14, 1029 (1965).

[4]F. Brown and R. E. Parks, Phys. Rev. Letters 16, 507 (1966).

[5]R. W. Terhune, Solid State Design 4, 38 (1963).

[6]P. S. Pershan, Phys. Rev. 130, 919 (1963).

[7]E. Adler, Phys. Rev. 134, A728 (1964).

[8]H. Cheng and P. B. Miller, Phys. Rev. 134, A683 (1964).

[9]S. S. Jha, Phys. Rev. Letters 15, 412 (1965), and Phys. Rev. 140, A2020 (1965); S. S. Jha and C. W. Warke, Phys. Rev. 153, 751 (1967), and 162, 854(E) (1967).

[10]N. Bloembergen and Y. R. Shen, Phys. Rev. 141, 298 (1966).

[11]N. Bloembergen and P. S. Pershan, Phys. Rev. 128, 606 (1962).

[12]P. D. Maker, in Physics of Quantum Electronics, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Company, Inc., New York, 1966), p. 60.

[13]H. Holloway, private communication.

[14]American Institute of Physics Handbook, edited by D. E. Gray (McGraw-Hill Book Company, Inc., New York, 1963), Sect. 6; H. R. Philipp and E. A. Taft, Phys. Rev. 113, 1002 (1959), and 120, 37 (1960); L. G. Schulz, J. Opt. Soc. Am. 44, 357 (1954); L. G. Schulz and F. R. Tangherlini, J. Opt. Soc. Am. 44, 362 (1954); International Critical Tables (McGraw-Hill Book Company, Inc., New York, 1929), Vol. 5, p. 249.

BINDING ENERGY OF O^{16} WITH A VELOCITY-DEPENDENT NUCLEON POTENTIAL*

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The binding energy and single-particle energies have been calculated for O^{16} using reaction matrix theory and a meson-theoretical nucleon potential due to Green and Sawada. The binding obtained is 7.2 MeV/nucleon after Coulomb and center-of-mass corrections, in reasonable agreement with the experimental 7.98 MeV/nucleon.

Recent developments in the reaction-matrix (K-matrix) theory of the nuclear many-body problem indicate that potential energies in the particle spectrum should be neglected when defining the K matrix.[1] The binding energy is then calculated with good accuracy to first order in the K matrix thus defined.

Hard-core potentials like the Brueckner-Gammel-Thaler (BGT) or Hamada-Johnston (HJ) potentials then give much too little binding for nuclear matter[2] or for O^{16}.[3-6] Softening of the core, while preserving the free-scattering properties, increases the binding somewhat but, as it seems, not sufficiently. The static hard-core and soft-core potentials currently in use reproduce two-body scattering data quite well but are mainly phenomenological. In this Letter we report on some calculations using one of the meson-theoretical potentials derived by Green and coworkers.[7,8] The "one-parameter, almost relativistic, regularized potential" of Green and Sawada[8] (to be referred to as G.S. 1) was chosen for simplicity.

The method of calculating the K-matrix elements has been presented in previous papers.[4-6] In Fig. 1, curve A, the results are shown for the energy per particle of O^{16} for the potential used. There is no sign of saturation for reasonable values of the oscillator parameter β ($\hbar\omega = \beta\hbar^2/M$). However, the G.S. 1 potential that we used reproduces all but the $^1P_1$ phase shifts quite well. The total contribution of $^1P_1$ K-matrix elements to the energy of O^{16} is shown in Fig. 2 in the curve labeled "G.S. 1." This contribution is positive and decreases with increasing β. For comparison is shown the corresponding $^1P_1$ contribution to the energy for the HJ potential. The latter is seen to increase quite