

FIG. 2. Schematic drawing of R rays after Müstel.⁸

tober may have been a direct consequence of the filamentary structure of the rays.

The second of the two events we wish to mention occurred on 1 December 1961. It is similar to that of 27 October, and the relevant sequence of events is also outlined in Table I. On 10 November 1961 a flare accompanied by Type-IV emission occurred on the extreme west limb of the sun and initiated a solar proton event which was observed by Explorer XII. No magnetic storm, Forbush decrease, or >3 -MeV proton increase was seen after the usual one- to three-day plasma transit time from the sun to the earth. Three weeks later on 1 December 1961, after $3/4$ of a solar rotation, there was a Forbush decrease, a magnetic storm, and a >3 -MeV proton event similar to that of 27 October, indicating the arrival of a solar plasma stream. The occurrence of the event of 1 December closely coincided with the central meridian passage of the active region which produced the flare on 10 November. This event again substantiates the picture of a long-lived plasma stream emanating from an active region of the sun, but in this case

the timing was different owing to the different position of the parent flare on the sun.

There is another occurrence which could be the same phenomenon. On 7 September 1961 a solar proton event having an anomalously slow intensity decay was observed by Explorer XII, the details of which will be reported in a later paper. There was no observation at that time of a large flare or of Type-IV emission which can definitely be associated with this event, and there was no geophysical disturbance two days later. Eleven days later on 18 September there was a small increase of the intensity of >3 -MeV protons similar in spectrum to those of 27 October and 1 December and unaccompanied by a flare. We speculate from these observations that the flare responsible for the solar proton event of 7 September occurred on the remote side of the sun and that the increase on 18 September was the same phenomenon as that observed on 27 October and 1 December.

*NAS-NASA Resident Research Associate.

¹S. Chapman and V. C. A. Ferraro, *Nature* **126**, 129 (1930).

²J. Bartels, *Terrest. Magnetism Atm. Elec.* **37**, 48 (1932).

³C. W. Snyder and M. Naugebauer, Fourth International Space Science Symposium, COSPAR, Warsaw, Poland, 1963 (unpublished).

⁴D. A. Bryant, T. L. Cline, U. D. Desai, and F. B. McDonald, *J. Geophys. Res.* **67**, 4983 (1962).

⁵E. N. Parker, *Space Science Reviews* **1**, 62 (1962).

⁶T. Gold, *J. Geophys. Res.* **64**, 1665 (1959).

⁷E. N. Müstel, *Astron. Zh.* **39**, 418 (1962) [translation: *Soviet Astron.-AJ* **6**, 333 (1962)].

⁸E. N. Müstel, *Astron. Zh.* **39**, 619 (1962) [translation: *Soviet Astron.-AJ* **6**, 488 (1963)].

QUANTITATIVE STUDIES OF OPTICAL HARMONIC GENERATION IN CdS, BaTiO₃, AND KH₂PO₄ TYPE CRYSTALS

R. C. Miller, D. A. Kleinman, and A. Savage

Bell Telephone Laboratories, Murray Hill, New Jersey

(Received 2 July 1963)

Recent theoretical discussions¹⁻⁴ of second-harmonic generation (SHG) have raised two questions which can be answered only by quantitative measurements of the second-order polarization tensor d . The first question is whether or not the d tensor obeys the symmetry proposed by Kleinman,² which would follow from the general symmetry derived by Armstrong et al.⁵ if the nonlinear mechanism is lossless and dis-

persionless. The second question is whether or not large values of d result when the second-harmonic frequency ν_2 approaches the intrinsic absorption edge of the crystal as proposed by Lax et al.^{3,4} The crystals chosen for this quantitative study are among the strongest known for SHG.⁶

We have measured the allowed components $d_{14} = d_{25}$ and d_{36} for KH₂PO₄, KD₂PO₄, and NH₄H₂PO₄

Table I. Summary of second-harmonic generation experiments giving the tensor d and the coherence length l_{coh} as defined in reference 12. For each laser d values are relative to $d_{36}=1.00$ for KH_2PO_4 . The calculated coherence lengths l_{calc} are given where refractive index data were available.

Crystal	Laser	d	l_{coh}	l_{calc}
KH_2PO_4	Ruby	$d_{36}=1.00$	18.5μ	18.8μ
		$d_{14}=0.95 \pm 0.06$	7.3	7.2
KD_2PO_4	Ruby	$d_{36}=0.75 \pm 0.02$	20.6	...
		$d_{14}=0.76 \pm 0.04$	7.7	...
$\text{NH}_4\text{H}_2\text{PO}_4$	Ruby	$d_{36}=0.93 \pm 0.06$	17.7	18.2
		$d_{14}=0.89 \pm 0.04$	6.7	6.4
KH_2PO_4	Nd	$d_{36}=1.00$	22.0	22.0
		$d_{14}=1.01 \pm 0.05$	14.6	15.0
KD_2PO_4	Nd	$d_{36}=0.92 \pm 0.04$	21.2	...
		$d_{14}=0.91 \pm 0.03$	15.8	...
$\text{NH}_4\text{H}_2\text{PO}_4$	Nd	$d_{36}=0.99 \pm 0.06$	21.0	20.6
		$d_{14}=0.98 \pm 0.05$	13.2	13.0
CdS	Nd	$d_{15}=35 \pm 2$	1.8	1.9
		$d_{31}=32 \pm 2$	1.7	1.6
		$d_{33}=63 \pm 4$	1.8	1.7
BaTiO_3	Nd	$d_{15}=35 \pm 3$	3.1	...
		$d_{31}=37 \pm 3$	5.8	...
		$d_{33}=14 \pm 1$	4.1	...

using the ruby laser ($h\nu_2=3.574$ eV). Results are given in Table I relative to d_{36} for KH_2PO_4 . It is found that in each case the proposed symmetry² $d_{14}=d_{25}=d_{36}$ is obeyed within experimental accuracy. It should be cautioned, however, that only the magnitude of d is measured in these experiments. Previously⁷ it has been shown that quartz obeys the above symmetry, which in that case reduces to $d_{14}=0$. Using the $\text{CaWO}_4:\text{Nd}^{+3}$ laser ($h\nu_2=2.343$ eV), we have measured the allowed components of d for KH_2PO_4 , CdS, and BaTiO_3 relative to d_{36} for KH_2PO_4 . In CdS and BaTiO_3 the proposed symmetry is $d_{15}=d_{31}$, which is found to be obeyed within experimental accuracy. It may be noted that the other component d_{33} in each case is distinctly different in magnitude. It is of practical interest that d_{33} for CdS is 63 times larger than d_{36} for KH_2PO_4 .

The effect of the intrinsic absorption edge on the magnitude of d was studied in CdS. The edge⁸ at room temperature lies at 2.48 eV, only 0.14 eV above $h\nu_2$ for the Nd laser. By raising the temperature the edge can be lowered until it coincides with $h\nu_2$ at about 235°C. We

have measured the SHG in CdS as a function of temperature in the range 25-300°C. The results are shown by the stars in Fig. 1 for an experimental arrangement which measures d_{33} . The open circles across the top give the laser intensity, which was monitored throughout the experiment and remained substantially constant. The scatter of points below 125°C is due to oscillations of the SHG as the refractive index changes with temperature. Above 125°C the SHG is substantially constant with no evidence of any rise around 235°C. It could not be determined with certainty whether d_{33} rises or falls with increasing temperature, but there is some indication that d_{33} falls gradually with temperature above 100°C. The effect of temperature was also studied in BaTiO_3 , where it was observed that d_{31} decreases with increasing temperature faster than the spontaneous polarization,⁹ and drops discontinuously to zero at the Curie temperature 120°C.

An interesting effect was observed in BaTiO_3 during ferroelectric polarization reversal. It was found that reversal enhanced the SHG by about one order of magnitude. This effect is believed to be due to the antiparallel domains formed during reversal.¹⁰ It can be shown that a single 180° domain wall suitably placed can increase SHG by a factor of four. The effect was also observed in triglycine sulfate.

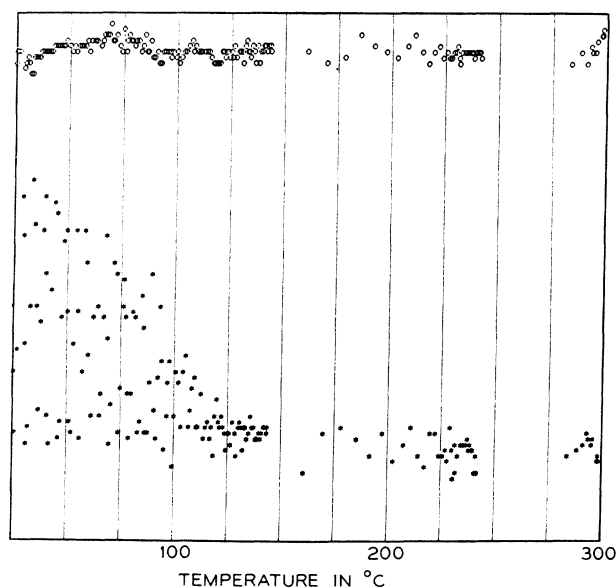


FIG. 1. SHG in CdS (stars) as a function of temperature °C together with the monitor (open circles at top) of the laser pulses.

Another interesting effect observed here in CdS and BaTiO₃¹¹ with the ruby laser is SHG under conditions in which the crystal is opaque to light of the second-harmonic frequency ν_2 . Outputs were observed comparable to those produced by a (110) plate of KH₂PO₄, which is not opaque at ν_2 . SHG in the opaque case was also observed in GaAs with the Nd laser. The statement by Kleinman¹² that absorption at ν_2 would prevent observation of SHG is therefore incorrect.

All measurements were made using polarized unfocused laser beams, a photomultiplier detection system responding only to the second harmonic and equipped with an analyzer, and samples in the form of plates with flat parallel faces of good optical quality. In this experiment^{13,14} one observes the interference between the forced wave produced by the laser beam and the free wave produced at the surface.¹² The coherence length can be determined from the oscillations of the SHG with respect to the angle of incidence of the beam. By suitable choices of polarizations and crystal orientation, each component of d could be measured separately. The relative magnitudes of the d components were determined by extrapolating the peaks of the oscillations to normal incidence. The dependence of the SHG upon the refractive indices was fully taken into account. The values obtained for the coherence lengths are listed in Table I along with values calculated from refractive index data. The measured and calculated values are seen to be in satisfactory agreement.

The measurements on CdS and BaTiO₃ as a function of temperature also give oscillations due to the change of the refractive index at frequency ν_2 with temperature. The oscillations damp out with increasing temperature, as seen in Fig. 1, due to increased absorption of the free wave. The forced wave cannot be absorbed, however, since it is continuously generated by the laser beam. This explains SHG in the opaque case. The oscillations are clearly seen in Fig. 2, where the points of Fig. 1 in the range 25-75°C are replotted on an expanded temperature scale. The curve is a theoretical^{12,15} fit taking into account the changing refractive index and absorption coefficient as well as a small amount of incoherence arising presumably from imperfection of the crystal surface. The value obtained for the absorption coefficient at 113°C is 83 cm⁻¹ in good agreement with an extrapolation of Dut-

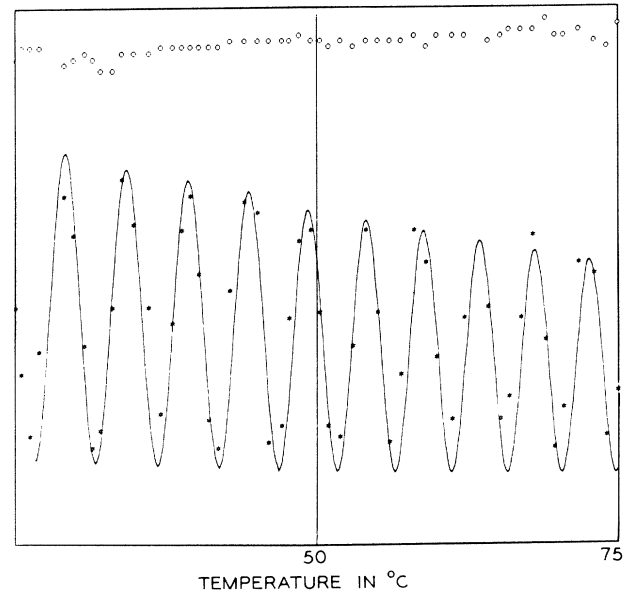


FIG. 2. The data of Fig. 1 from 25-75°C together with a theoretical fit (curve) showing the oscillatory behavior.

ton's⁸ measurements.

Recently Ashkin, Boyd, and Dziedzic¹⁶ have measured d_{36} for KH₂PO₄ using the continuous He-Ne gas laser ($h\nu_2 = 2.151$ eV). We may infer² from the fact that the symmetry $d_{14} = d_{36}$ is obeyed in KH₂PO₄ that dispersion in d is very small. The values of Table I are then put on an absolute basis by adopting the value of Ashkin, Boyd, and Dziedzic¹⁶ $d_{36} = (3 \pm 1) \times 10^{-9}$ esu (KH₂PO₄).

¹P. A. Franken and J. F. Ward, Rev. Mod. Phys. **35**, 23 (1963).

²D. A. Kleinman, Phys. Rev. **126**, 1977 (1962).

³B. Lax, J. G. Mavroides, and D. F. Edwards, Phys. Rev. Letters **8**, 166 (1962).

⁴R. Loudon, Proc. Phys. Soc. (London) **80**, 952 (1962).

⁵J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127**, 1918 (1962).

⁶A. Savage and R. C. Miller, Appl. Opt. **1**, 661 (1962).

⁷Robert C. Miller, Phys. Rev. **131**, 95 (1963).

⁸David Dutton, Phys. Rev. **112**, 785 (1958). The absorption edge is here defined as the sharp knee at which the absorption coefficient turns downward with decreasing energy. A review of data on CdS is given by T. S. Moss, *Optical Properties of Semiconductors* (Butterworths Scientific Publications, Ltd., London, 1959), Sec. 15.6.

⁹See, for example, F. Jona and G. Shirane, *Ferro-*

electric Crystals (The Macmillan Company, New York, 1962), p. 115.

¹⁰Domain effects have also been reported for KH_2PO_4 by J. Van der Ziel and N. Bloembergen, *Bull. Am. Phys. Soc.* **8**, 380 (1963).

¹¹F. Brown, *Bull. Am. Phys. Soc.* **8**, 62 (1963).

¹²D. A. Kleinman, *Phys. Rev.* **128**, 1761 (1962).

¹³P. D. Maker, R. W. Terhune, M. Nisenoff, and C. M. Savage, *Phys. Rev. Letters* **8**, 21 (1962).

¹⁴J. A. Giordmaine, *Phys. Rev. Letters* **8**, 19 (1962).

¹⁵N. Bloembergen and P. S. Pershan, *Phys. Rev.* **128**, 606 (1962).

¹⁶A. Ashkin, G. D. Boyd, and J. M. Dziedzic, *Phys. Rev. Letters* **11**, 14 (1963).

INFLUENCE OF TRANSPORT CURRENT ON THE MAGNETIZATION OF A HARD SUPERCONDUCTOR*

M. A. R. LeBlanc†

Stanford University, Stanford, California

(Received 7 June 1963)

The influence of transport current on the magnetization of cold-worked Nb-Zr wire in a transverse field has been studied at 4.2°K. The magnetic moment (diamagnetic or paramagnetic) is reduced by the transport current and tends to zero as the critical current is approached.¹ At any given field the curve of magnetic moment versus transport current is not single-valued, but is seen to depend on the sequence of application of the current and field and also on the history of the transport current at the final field. The results can be understood on the basis of the Bean model for the magnetization of a hard superconductor² when the field produced by the transport current is taken into consideration.

The sample consists of a noninductive, close-wound, single-layer coil, 3 cm in length, 1.4 cm in diameter, of severely cold-worked, Formvar insulated, 10-mil diameter Nb-Zr (25%) wire. The magnetization of the sample is determined by ballistically measuring the emf induced in a pick-up coil surrounding the sample when the latter is suddenly driven well above its transition temperature by a heat pulse.

Curve C of Fig. 1 shows the initial magnetization as the field is increased, and curve F gives the paramagnetic moment vs final field after cooling in a field of 12 kG. The maximum magnetization in a field H_a is independent of previous history provided the final change of the applied field which induces the magnetization is sufficient to achieve saturation of the flux screening or flux trapping currents at the final field. Curves D and E of Fig. 1 show the approach to saturation magnetization for different initial conditions, i.e., cooling in 4.0 and 6.3 kG, respectively. No evidence of a Meissner effect upon cooling in a field was observed at any field.

Curve A of Fig. 1 gives the critical current I_c vs H_a . This critical current is reproducible

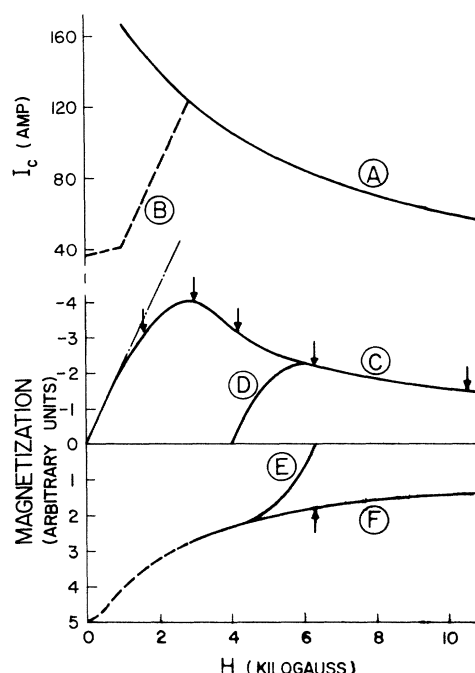


FIG. 1. Curve A—Critical current vs transverse external field. Curve B—Critical current for initial resistive transition vs external field for paramagnetic critical state. Curve C—Initial magnetization. Curves D and E—Approach to saturation magnetization after cooling in 4.0 and 6.3 kG, respectively. Curve F—Saturation paramagnetic moment vs external field after cooling in 12 kG.

(no training occurs) and independent of the sequence of application of field and current and of the magnetization, provided the latter lies on curve C and the solid part of curve F or inside the region defined by these and the horizontal axis. Curve B gives the critical current for the initial resistive transitions (approximately independent of the sequence of application of cur-