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Laser-Induced Kerr Constants for Pure Liquids

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During the past two decades an increasing number of publications have reported laser-induced birefringence data for pure liquids. To date there has been no comparative collection of values from these experiments. This paper lists the published values together with hitherto unpublished data of the authors of the optically induced Kerr constant B_0. The normalized parameter B_{rel} for data compared with benzene under stated wavelength conditions of the light used for the measurements is also given.

Key words: Kerr constants; Kerr effect; laser-induced phenomena; nonlinear optics; optical birefringence; pure liquids.

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1. Introduction

When an electric field is applied to an isotropic medium it induces changes in the refractive index of the medium. This is the "Kerr effect." With the advent of high-power lasers, a new series of experiments has evolved in which the electric vector in the laser beam is of sufficient amplitude to induce the refractive index changes. The vectorial nature of the field results in the directional changes in the active medium which are detected in terms of the induced optical birefringence (Δn). This optically induced Kerr effect (OKE) was predicted by Buckingham,[1] and was first observed for liquids by Mayer and Gires[2] in 1964, and by Wong and Shen[3] (1973) for liquid crystals. This review is concerned only with pure liquids.

During the past few years a number of experiments have been carried out to determine the Optical Kerr constants of various pure liquids.[4–18] In addition, work has been published on molecular solutions,[19,20] colloidal and macromolecular suspensions,[21,22] liquid crystals,[23,24] and solids.[10,25] Three methods have been used to quantify the effect. These are (a) measurement of the linearly induced birefringence (this is the original Kerr effect); (b) measurement of the changes in the ellipticity of incident, elliptically polarized light; and (c) detection of refractive index changes via interferometric methods. With fluid media, the phenomenon is associated with field-induced molecular reorientation.[26] By far the majority of experiments have been based on birefringence measurements.

As the phenomenon relates directly to the third-order nonlinear susceptibility of an active medium, and because of the potential of methods for the basis of fast, optically activated switching and modulation mechanisms, there is a growing interest in the effect. To date, however, there is no comparative collation of the reported data for the pure liquids studied. This review seeks to redress this imbalance and to provide a compendium of related magnitudes for the liquids. Restriction is made to birefringence data induced by nanosecond laser pulses.

2. Background Theory

When an intense monochromatic laser beam of wavelength λ_i traverses an isotropic medium, it induces a nonlinear polarization (P) of the form:[27]

$$P_i^{(3)} = \Sigma_{jkl}\chi_{ijkl}^{(3)}(-\omega,\omega,-\omega,\omega) \times E_j(\omega)E_k(\omega)E_l^{*}(\omega) \qquad (1)$$

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where ω is the inducing beam frequency, $\chi^{(3)}_{ijkl}$ is the third-order nonlinear susceptibility tensor of the medium and E is the electric vector amplitude. For an isotropic pure liquid, the only independent nonzero components[28] of the tensor $\chi^{(3)}_{ijkl}$ are $\chi^{(3)}_{1122}$, $\chi^{(3)}_{1212}$, and $\chi^{(3)}_{1221}$, whilst

$$\chi^{(3)}_{1111} = \chi^{(3)}_{1122} + \chi^{(3)}_{1212} + \chi^{(3)}_{1221}. \qquad (2)$$

The significance of this equation has been given by Shen.[27]

The Optical Kerr effect is usually studied by measuring the linear birefringence induced by the laser beam in the active medium. This is accomplished by sending a second low-intensity probe laser beam of wavelength λ_p colinear to the inducing beam through the medium. The inducing and probe beams are both linearly polarized and adjusted so that the probe beam polarization azimuth is set at 45° to that of the inducing beam. Since the inducing laser produces a difference in refractive index within the liquid for light polarized parallel and perpendicular to the polarization azimuth of the inducing beam, this has the effect of changing the relative phase of the two components of the probe beam linearly resolved along these same directions. Hence the probe beam becomes elliptically polarized, and the induced birefringence Δn can be measured. Reference 29 reviews the optical arrangements used to date and Ref. 30 describes birefringence detection procedures.

By considering the polarization states of the inducing and probe beams it can be shown that[27]

$$\Delta n(\omega) = (2\pi/n)(\chi^{(3)}_{1212} + \chi^{(3)}_{1221})|E(\omega)|^2. \qquad (3)$$

Conventionally, for pure liquids the Kerr constant B has been measured where

$$B = \Delta n/\lambda_p |E|^2. \qquad (4)$$

In this work, the zero subscript indicates the laser-induced effect. Hence,

$$B_0 = (2\pi/n\lambda_p)(\chi^{(3)}_{1212} + \chi^{(3)}_{1221}). \qquad (5)$$

From this equation it is seen that measurements of Δn, and hence B_0, at the high frequencies of an optical beam cannot be transposed into $\chi^{(3)}$ data unless the interrelation between χ^3_{1212} and χ^3_{1221} is known. The corollary is also true in that specific components of $\chi^{(3)}$ must be measured (or inferred from molecular structure) if B_0 is to be enumerated from $\chi^{(3)}$ data. Since the majority of experiments on pure liquids have involved the direct measurement of the laser-induced birefringence, it is these results that are summarized here.

In almost all OKE experiments the inducing beam has been a "Q"-switched pulse from a solid-state laser. The only exception for pure liquids has been the recent work of Buckingham and Williams[16] who used a weakly focused argon-ion laser. One of the major difficulties in OKE experiments has been the calculation of the electric field in the inducing laser pulse. Many different formulas have been used,[13,17,18,21] some of which have taken absorption effects[13,18] into account. To overcome this problem it has become a common practice to report data relative to values of B_0 measured for benzene or carbon disulphide.

This paper presents a collation of all reported values of Optical Kerr constants for pure liquids. The authors' original values of B_0 are given. As the majority of studies have included measurements on benzene, we have calculated values of $B_{\rm rel}$, the Kerr constants for each liquid relative to the same authors' value for benzene. The authors' own estimates of uncertainty have been indicated in Table 1 as $\Delta B_{\rm rel}$. In Table 1 all Kerr constants are given in S. I. units. To convert to e.s.u. units, one must multiply the tabulated values by 8.988×10^6.

3. Discussion

From Table 1, it is apparent that, for certain liquids, the values of B_0 reported by different authors show a wide variance that is in excess of their stipulated experimental errors. It is noteworthy, however, that this disparity between the reported results is generally significantly reduced when data are compared for $B_{\rm rel}$, provided that the authors' own original values for benzene are used. Hence, the somewhat low absolute values of Ref. 14 or high values of Ref. 11 give rise to typical data for the Optical Kerr constants when normalized to benzene. Only in the case of Gires[9] is this not true. His consistently high values for $B_{\rm rel}$ may reflect his low value of B_0 for benzene which might have been related to sample impurity. The concord in $B_{\rm rel}$ values is comforting in that it demonstrates comparative consistency between sets of data. It does indicate a significant difference of approach by the various experiments in their analytical procedures for transposing experimental data into absolute values of B_0.

A variety of reasons can contribute to this uncertainty, as has been briefly discussed elsewhere.[29] Both experimental and theoretical factors are of relevance here. Experimentally, there was a failure in early measurements to use strictly monomodal lasers for the inducing beam. As was pointed out by Jennings and Coles,[21] only this configuration has a unidirectional E vector in the beam. Other configurations may result in overestimation of the effective E with consequential reflection in the calculated values of B_0. A further experimental complication arises from a failure to take account of the spatial intensity profile of the laser beams. This was pointed out by Pouligny *et al.*[31] In a laser beam the field E is not uniform in space. The unmatched diameters of the probe and inducing beams used by Coles and Jennings may account for their low values of B_0 for various liquids reported in Ref. 14. Complications in the estimation of E also arise from the use of different theoretical expressions for this parameter as mentioned in the Introduction. The various experimental optical configurations, with different mode structures, beam profiles, and the use of focused or parallel optical beams are all matched with different theoretical expressions for the estimation of E. In a number of cases, no relevant equation is given, and discrepancies in absolute values for B_0 result.

In the present authors' view, this uncertainty in measurement of the laser field E is the most significant hindrance to establishing reliable absolute values of the Optical Kerr constant. Until experimental consistency evolves, the use of $B_{\rm rel}$ values is recommended for interlaboratory comparisons.

Such a recommendation focuses on the need for a "best" value for the Optical Kerr constant for benzene.

TABLE 1. Optical Kerr constants for pure liquids. (ΔB_{rel} values are the authors' quoted uncertainties in B_{rel}.)

Pure liquid	B_{rel}	$B_0\times10^{-14}$ $V^{-2}m$	ΔB_{rel} (%)	λ_i (nm)	λ_p (nm)	Reference
Acetic acid	0.327	0.147	–	694	488	11
	0.422	0.217	5	1064	442	18
	0.418	0.331	5	532	442	18
Acetone	0.18	0.081	–	694	488	11
Acetophenone	–	1.68	>8	1060	633	17
Anethole	9.83	6.9	5	694	488	7
Anisole	1.44	1.01	5	694	488	7
Benzene	1	0.7015	5	694	488	4
	1	0.577	20	694	488	6
	1	0.445	–	694	488	9
	1	0.45	–	694	488	11
	1	0.78	–	694	500	2
	1	0.1	30	1064	488	14
	1	0.514	5	1064	442	18
	1	0.791	5	532	442	18
Benzoyl chloride	4.7	3.67	–	694	500	2
	3.0	0.3	20	1064	488	14
	3.94	2.03	5	1064	442	18
p-Bromoanisole	3.72	2.61	5	694	488	7
o-Bromoanisole	1.93	1.35	5	694	488	7
Bromobenzene	1.97	–	30	1060	633	15
	–	2.03	>8	1060	633	17
Bromonaphthalene	–	2.60	>8	1060	633	17
m-Bromotoluene	2.33	1.34	20	694	488	6
Butyric acid	0.357	0.184	5	1064	442	18
	0.384	0.304	10	532	442	18
Carbon disulphide	6.01	3.47	10	694	488	5
	10.5	4.67	–	694	488	9
	10.3	4.65	–	694	488	11
	5.56	4.34	–	694	500	2
	7.1	–	–	694	442	12
	6.6	–	–	1060	633	12
	6.9	–	–	1060	530	12
	7.3	0.73	14	1064	488	14
	3	–	17	488	633	16
Carbon tetrachloride	0.127	0.057	–	694	488	11
	0.065	–	–	1060	530	12
Chlorobenzene	1.76	–	30	1060	633	15
	–	1.26	>8	1060	633	17
Chloroform	0.24	0.138	10	694	488	5
	0.409	0.184	–	694	488	11
	0.35	–	–	1060	633	12
α-Chloronaphthalene	6.76	3.04	–	694	488	11
o-Chlorotoluene	2.46	1.42	20	694	488	6
p-Chlorotoluene	2.24	1.29	20	694	488	6
2,4,6-Collidine	2.69	1.55	20	694	488	6
Cyclohexane	0.09	0.052	10	694	488	5
	0.095	0.067	5	694	488	7
	0.1025	0.046	–	694	488	9
	0.1022	0.046	–	694	488	11
Cyclohexanone	0.181	0.127	5	694	488	7
Cyclooctane	0.109	0.0765	5	694	488	7
Decane	0.194	0.136	5	694	488	4
	0.206	0.106	10	1064	442	18

TABLE 1. Optical Kerr constants for pure liquids. (ΔB_{rel} values are the authors' quoted uncertainties in B_{rel}.)—Continued

Pure liquid	B_{rel}	$B_0 \times 10^{-14}$ $\mathrm{V^{-2}m}$	ΔB_{rel} (%)	λ_i (nm)	λ_p (nm)	Reference
1,9-Decadiene	0.374	0.192	5	1064	442	18
1-Decanol	0.207	0.106	5	1064	442	18
1-Decene	0.323	0.166	5	1064	442	18
	0.33	0.257	10	532	442	18
1-Decyne	0.346	0.178	5	1064	442	18
Deuterated water	0.057	0.029	5	1064	442	18
Dibromobenzene	2.25	–	30	1060	633	15
o-Dichlorobenzene	–	1.91	>8	1060	633	17
	2.56	1.48	20	694	488	6
	2.56	–	30	1060	633	15
m-Dichlorobenzene	2.43	1.4	20	694	488	6
	2.43	–	30	694	488	15
Dodecane	0.220	0.154	5	694	488	4
	0.233	0.120	5	1064	442	18
1-Dodecanol	0.250	0.129	5	1064	442	18
1-Dodecene	0.30	0.219	5	532	442	18
	0.28	0.179	5	1064	442	18
1-Eicosene	0.367	0.189	5	1064	442	18
Ethanol	0.100	0.051	5	1064	442	18
Formic Acid	0.482	0.248	10	1064	442	18
Heptane	0.162	0.114	5	694	488	4
	0.149	0.077	5	1064	442	18
Heptanoic acid	0.378	0.194	5	1064	442	18
1-Heptanol	0.159	0.082	5	1064	442	18
1-Heptene	0.332	0.171	10	1064	442	18
	0.36	0.283	5	532	442	18
1-Heptyne	0.374	0.192	10	1064	442	18
Hexadecane	0.288	0.202	5	694	488	4
1-Hexadecene	0.391	0.201	5	1064	442	18
1,5-Hexadiene	0.572	0.294	10	1064	442	18
Hexane	0.142	0.100	5	694	488	4
	0.136	0.070	5	1064	442	18
Hexanoic acid	0.372	0.191	5	1064	442	18
1-Hexanol	0.136	0.070	5	1064	442	18
1-Hexene	0.287	0.148	10	1064	442	18
	0.31	0.242	5	532	442	18
1-Hexyne	0.359	0.183	10	1064	442	18
2,6-Lutidine	1.54	0.885	20	694	488	6
3,4-Lutidine	1.83	1.05	20	694	488	6
2,4-Lutidine	2.47	1.42	20	694	488	6
Mesitylene	1.6	–	–	694	442	12
	1.5	–	–	1060	633	12
Methanol	0.065	0.034	5	1064	442	18
Methyl cyanide (acetonitrile)	0.43	0.248	–	694	488	5
Methyl cyclooctane	0.105	0.0737	5	694	488	7
Methyl naphthalene	–	1.78	>8	1060	633	17

TABLE 1. Optical Kerr constants for pure liquids. ($\Delta B_{\rm rel}$ values are the authors' quoted uncertainties in $B_{\rm rel}$.)—Continued

Pure liquid	$B_{\rm rel}$	$B_0 \times 10^{-14}$ $\rm V^{-2}m$	$\Delta B_{\rm rel}$ (%)	λ_i (nm)	λ_p (nm)	Reference
Methylocyclohexane	0.11	0.063	10	694	488	5
o-Nitroanisole	2.76	1.94	5	694	488	7
	3.3	–	–	1060	633	13
Nitrobenzene	7.25	3.23	–	694	488	9
	4.71	3.67	–	694	500	2
	5.3	–	–	1060	633	12
	4.2	–	–	1060	633	13
	6.3	0.63	14	1064	488	14
	–	2.63	>8	1060	633	17
	4.75	2.44	10	1064	442	18
o-Nitrotoluene	3.1	1.79	20	694	488	6
m-Nitrotoluene	2.9	1.67	20	694	488	6
p-Nitrotoluene	2.64	1.52**	20	694	488	6
Nonane	0.183	0.128	5	694	488	4
	0.170	0.087	5	1064	442	18
Nonanoic acid	0.395	0.203	5	1064	442	18
	0.295	0.233	5	532	442	18
1-Nonanol	0.197	0.101	5	1064	442	18
1-Nonene	0.333	0.171	5	1064	442	18
	0.37	0.292	5	532	442	18
1-Nonyne	0.397	0.204	10	1064	442	18
1-Octadecene	0.400	0.206	5	1064	442	18
1,7-Octadiene	0.542	0.279	5	1064	442	18
Octane	0.173	0.121	5	694	488	4
	0.171	0.088	5	1064	442	18
Octanoic acid	0.393	0.202	5	1064	442	18
	0.315	0.249	5	532	442	18
1-Octanol	0.178	0.092	5	1064	442	18
1-Octene	0.290	0.149	5	1064	442	18
	0.43	0.344	10	532	442	18
1-Octyne	0.392	0.201	5	1064	442	18
Pentadecane	0.263	0.184	5	694	488	4
Pentane	0.125	0.088	5	694	488	4
	0.13	0.075	10	694	488	5
	0.102	0.052	5	1064	442	18
1-Pentanol	0.121	0.062	5	1064	442	18
1-Pentene	0.332	0.171	5	1064	442	18
2-Pentene	0.37	0.213	10	694	488	5
1-Pentyne	0.432	0.222	10	1064	442	18
Phosyl chloride	–	0.226	20	1060	530	10
α-Picoline	1.56	0.90	20	694	488	6
β-Picoline	1.79	1.02	20	694	488	6
γ-Picoline	2.08	1.20	20	694	488	6
1-Propanol	0.101	0.052	5	1064	442	18
Propionaldehyde	0.183	0.081	–	694	500	9
Propionic acid	0.403	0.207	5	1064	442	18
	0.384	0.304	10	532	442	18
Pyridine	1.38	0.795	20	694	488	6
	1.5	0.15	27	1064	488	14

TABLE 1. Optical Kerr constants for pure liquids. (ΔB_{rel} values are the authors' quoted uncertainties in B_{rel}.)—Continued

Pure liquid	B_{rel}	$B_0 \times 10^{-14}$ $V^{-2}m$	ΔB_{rel} (%)	λ_i (nm)	λ_p (nm)	Reference
Tetrachloroethylene	2.11	1.22	10	694	488	5
Tetradecane	0.253	0.177	5	694	488	4
1-Tetradecene	0.33	0.170	10	1064	442	18
	0.39	0.307	10	532	442	18
Toluene	1.32	0.796	10	694	488	5
	1.49	0.860	20	694	488	6
	2.33	1.04	–	694	500	9
	–	1.05	20	1060	530	10
	2.29	1.03	–	694	488	11
	1.3	–	–	694	442	12
	1.4	–	–	1060	633	12
	1.4	–	–	1060	633	13
	1.6	0.16	25	1064	488	14
	1.25	0.64	10	1064	442	18
Trichloroethylene	2.4	1.39	10	694	488	5
1,1,2-Trichloro-1,2,3-trifluoroethane	0.25	–	–	1060	530	12
1-Tridecene	0.32	0.252	15	532	442	18
Undecane	0.208	0.146	5	694	488	4
	0.211	0.108	10	1064	442	18
1-Undecanol	0.218	0.112	5	1064	442	18
1-Undecene	0.324	0.167	10	1064	442	18
	0.30	0.239	10	532	442	18
Valeric acid	0.310	0.245	5	532	442	18
	0.380	0.195	5	1064	442	18
Water	0.053	0.037	9	694	488	8
	0.073	0.032	–	694	500	9
	0.071	0.032	–	694	488	11
	0.040	0.021	5	1064	442	18
o-Xylene	1.85	1.07	20	694	488	6
m-Xylene	1.79	1.03	20	694	488	6
p-Xylene	1.72	0.99	20	694	488	6

**At 62 °C

From Table 1, the results may be classified into two broad groups of "high" and "low" values. No apparent logic dissociates the results between these groupings. Like refractive index, $\chi^{(3)}$ and hence B_0 must exhibit wavelength dispersion effects. This is not only in the wavelength dependence of the measured birefringence, but also through the variable frequency of the electric vector in the inducing beam. For $\chi^{(3)}$ this has been discussed by Levenson and Bloembergen[32] and Kuzyk *et al.*[33] For all reported values for benzene, probe beams in the blue/green spectral region have been used.

It is noted that both of the most recently reported sets of values (Refs. 4 and 18) are in the "high" group for visible wavelength inducing beams. They also correspond to the most sophisticated experimental arrangements used to date. Furthermore when plotted as a wavelength dispersion curve, the three values of B_0 fit on the curve shown in Fig. 1. The current authors' therefore suggest that, for blue/green measuring light, the most reliable optical Kerr constants are as follows for the stated inducing beam wavelengths:

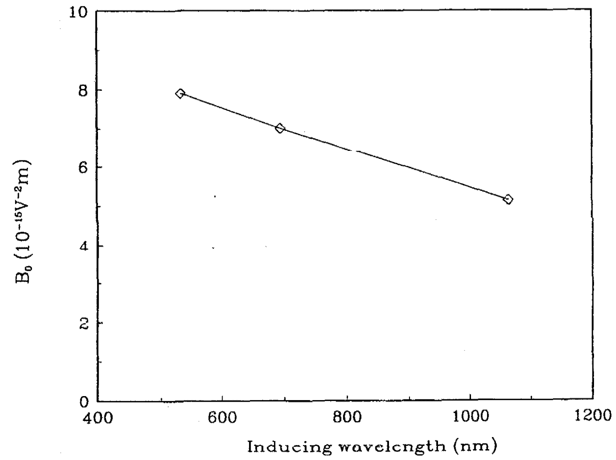

FIG. 1. The wavelength dispersion of B_0 for benzene with inducing beam wavelength. Data taken from Refs. 4 and 18.

$$B_0=0.51\times 10^{-14}\ \mathrm{V}^{-2}\,\mathrm{m}\ \text{at}\ 1064\ \text{nm},$$
$$B_0=0.70\times 10^{-14}\ \mathrm{V}^{-2}\,\mathrm{m}\ \text{at}\ 694\ \text{nm},$$
$$B_0=0.79\times 10^{-14}\ \mathrm{V}^{-2}\,\mathrm{m}\ \text{at}\ 532\ \text{nm}.$$

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