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ORIGINAL ARTICLE

Study of Photophysical Properties on Newly Synthesized Coumarin Derivatives

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Abstract Herein, we have studied the photophysical properties for three newly synthesized coumarin derivatives; 4-((2,6-dibromo-4-methylphenoxy)methyl)-2H-benzo[h]chromen-2-one (DMB), 4-((3,4-dihydro-6,7-dimethoxyisoquinolin-1-yl)methyl)-6-methyl-2H-chromen-2-one (DIM) and 4-((p-tolyloxy)methyl)-6-methoxy-2H-chromen-2-one (TMC). The absorption and emission spectra for above said molecules were recorded in different solvents at room temperature in order to calculate their ground and excited state dipole moments. The ground (μ_g) and excited state dipole (μ_e) moments of these coumarin derivatives were calculated using Lippert's, Bakshiev's and Kawski-Chamma-Viallet's equations by the solvatochromic shift method, which involves a variation of Stokes shift with the solvent dielectric constant and refractive index. Ground state dipole moments (μ_g) were also calculated from the Guggenheim method using the dielectric constant and refractive index of the solute molecule. The value of ground state dipole moment obtained from these two methods is well correlated. Further, it is notified that the excited state dipole moment is larger than the ground state dipole moment for all three solute molecules. It inferred that the excited state for above said molecules is more polar than the ground state. The present investigations may shine in the design of nonlinear optical materials.

Keywords Coumarin · Dipole moment · Electronic absorption spectra · Fluorescence spectra · Guggenheim method · Solvatochromic shift method

Introduction

Coumarin derivatives and analogues are well-known for their photochemical and photophysical properties as well as for their interesting second-order nonlinearities [1–6]. In addition, the coumarin unit is known to undergo a reversible photo-induced cyclodimerization by irradiation at λ > 300 nm that leads to stable cyclobutane based dimers, whereas the reverse photocleavage reaction occurs at shorter wavelengths (λ < 280 nm) [7–9]. Coumarin-derived Cu (II)-selective fluorescent sensor and studied the fluorescence quenching mechanism by femtosecond time-resolved fluorescence (TRF) spectroscopy and quantum calculations [10]. Coumarin derivatives studied to their widespread industrial use as dye lasers [11]. In recent years, there has also been a drive to synthesize coumarin-based organic dyes for use in high-efficiency dye-sensitized solar cells (DSCs) [12, 13]. Coumarin and its derivatives have attracted significant interest in a wide range of pharmaceutical research areas such as anti-inflammatory, hepatoprotective, antiviral, anticarcinogenic and anticoagulant activities [14–17] and they have various applications in food constituents, stabilizers and clinical use [18].

Knowledge of the excited state charge distribution and the dipole moments of the molecules are important in designing the nonlinear optical materials [19, 20] and understanding the photochemical processes. Methods available for the determination of dipole moments can be classified as either external or internal. External methods includes electric dichroism [21], Stark splitting of rotational levels [22]

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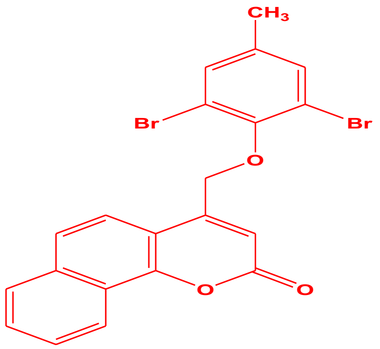
and microwave conductivity [23] are consider to be more accurate in the determining the excited state dipole moment of the simple organic molecules, however the quite popular internal method for the determination of the singlet excited state dipole moment is based on the analysis of absorption and fluorescence maxima. The procedure is evolved by Lippert's [24], Bakshiev's [25] and Kawski-Chamma-Viallet's equations [26] followed by the variation of Stokes shift with solvent polarity function.

In several investigations, they have reported regarding the estimation of ground and excited state dipole moments using various organic compounds [27–35]. However there is no report available in literature for the determination the ground and excited state dipole moments of these three 4-(2,6-Dibromo-4-methyl-phenoxymethyl)-benzo[h]chromen-2-one(DMB), 4-(6,7-Dimethoxy-3,4-dihydro-isoquinoline-1-ylmethyl)-6-methyl-chromen-2-one(DIM) and 4-((p-tolyloxy)methyl)-6-methoxy-2H-chromen-2-one (TMC) coumarin molecules. In this paper, we have estimated and compared the ground state and the excited state dipole moments of these three coumarin molecules by various methods.

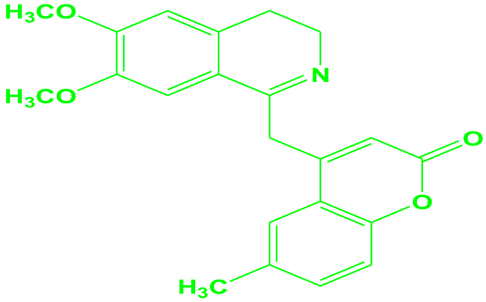
Materials and Methods

The novel coumarin derivatives namely; 4-((2,6-dibromo-4-methylphenoxy)methyl)-2H-benzo[h]chromen-2-one (DMB), 4-((3,4-dihydro-6,7-dimethoxyisoquinolin-1-yl)methyl)-6-methyl-2H-chromen-2-one (DIM) and 4-((p-tolyloxy)methyl)-6-methoxy-2H-chromen-2-one (TMC) were synthesised according to references [36, 37]. The molecular

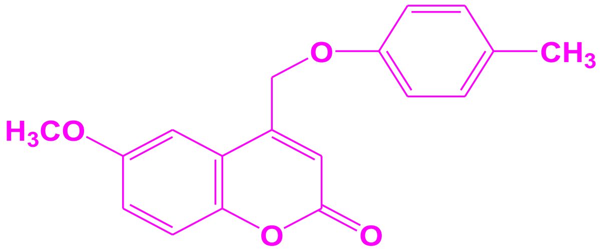
Fig. 1 The molecular structure of coumarin molecules (DMB, DIM and TMC)



4-((2,6-dibromo-4-methylphenoxy)methyl)-2*H*-benzo[*h*]chromen-2-one (DMB)



4-((3,4-dihydro-6,7-dimethoxyisoquinolin-1-yl)methyl)-6-methyl-2*H*-chromen-2-one (DIM)



4-((*p*-tolyloxy)methyl)-6-methoxy-2*H*-chromen-2-one (TMC)

structures of these three molecules are shown in Fig. 1. Solvents used in the present study were trichloroethylene, benzene, toluene, tetrahydrofuran, dioxin, acetone, acetonitrile, methanol and dimethyl sulfoxide [DMSO] were purchased from S-D Fine Chemicals Ltd. India and used without any further purification.

Several experimental techniques were used to characterize the coumarin molecules at room temperatures like Hitachi model U-3310 UV–visible spectrophotometer was used to record the electronic absorption spectra and Hitachi model F-7000 fluorescence spectrophotometer was used to record fluorescence spectra. The LCR Data Bridge was used to measure the dielectric constant and Abbe's refractometer was used to measure the refractive index of the coumarin molecules. All these measurements were carried out by keeping dye concentration very low about 1×10^{-5} M in order to avoid the self-absorption.

The dielectric constant of the solution ε_{xy} can be calculated using the equation

$$\varepsilon_{xy} = \frac{C_S - C_L}{C_A - C_L} \qquad (1)$$

where

C_S Capacitance of the solution.
C_A Capacitance of the air.
C_L Capacitance due to leads.

Solvatochromic Shift Method

The ground state and excited state dipole moments for all three molecules are calculated using three independent equations proposed by Lippert's, Bakshiev's and Kawaski-Chamma-Viallte's.

I. Lippert's equation [24] is given by

$$\overline{\upsilon_a} - \overline{\upsilon_f} = m\,\Phi(\varepsilon, n) + \text{Constant} \qquad (2)$$

Table 1 Spectral shift data for DMB molecule in different solvents

Solvent	$\overline{\upsilon_a}$ (cm^{-1})	$\overline{\upsilon_f}$ (cm^{-1})	$\overline{\upsilon_a} - \overline{\upsilon_f}$ (cm^{-1})	$\frac{\overline{\upsilon_a}+\overline{\upsilon_f}}{2}$ (cm^{-1})
Trichloroethylene	29325.51	23736.05	5589.95	26530.78
Benzene	29325.50	23752.96	5572.55	26539.35
Toluene	29325.51	23798.19	5527.32	26561.85
Dioxin	29411.76	23752.96	5658.80	26582.36
Tetrahydrofuran	29325.51	23501.76	5824.75	26413.35
Dimethyl sulphoxide	28985.50	22962.11	6023.11	25973.80
Acetone	29239.76	23228.80	6010.96	26234.28
Acetonitrile	29498.25	23375.40	6123.12	26436.96
Methanol	29498.52	23364.48	6134.04	26431.50

Table 2 Spectral shift data for DIM molecule in different solvents

Solvent	$\overline{\upsilon_a}$ (cm^{-1})	$\overline{\upsilon_f}$ (cm^{-1})	$\overline{\upsilon_a} - \overline{\upsilon_f}$ (cm^{-1})	$\frac{\overline{\upsilon_a}+\overline{\upsilon_f}}{2}$ (cm^{-1})
Trichloroethylene	28985.50	23640.66	5332.1	26279.9
Benzene	29411.76	24096.38	5321.6	26943.5
Toluene	29411.76	240673.8	5344.10	26739.10
Dioxin	29411.76	24061.59	5350.01	26693.18
Tetrahydrofuran	29325.51	23809.52	5512.12	26553.10
Dimethyl sulphoxide	29325.51	23781.21	5583.16	26320.50
Acetone	29325.51	23736.05	5589.13	26530.50
Acetonitrile	29585.79	23906.28	5662.02	26754.01
Methanol	29850.74	24154.58	5655.80	27022.50

II. Bakshiev's equation [25]

$$\overline{\upsilon_a} - \overline{\upsilon_f} = m_1\Phi_1(\varepsilon, n) + \text{Constant} \qquad (3)$$

III. Kawski-Chamma-Viallete's equation [26]

$$\frac{\overline{\upsilon_a} + \overline{\upsilon_f}}{2} = -m_2\Phi_2(\varepsilon, n) + \text{Constant} \qquad (4)$$

where $\Phi(\varepsilon, n) = \left[\frac{\varepsilon-1}{2\varepsilon+1} - \frac{n^2-1}{2n^2+1}\right]$ is the Lippert's polarity function.

$\Phi_1(\varepsilon, n) = \frac{2n^2+1}{n^2+2}\left[\frac{\varepsilon-1}{\varepsilon+2} - \frac{n^2-1}{n^2+2}\right]$ is the Bakshiev's polarity function.

$\Phi_2(\varepsilon, n) = \left[\frac{2n^2+1}{2(n^2+2)}\left(\left[\frac{\varepsilon-1}{\varepsilon+2} - \frac{n^2-1}{n^2+2}\right]\right) + \frac{3(n^4-1)}{2(n^2+2)^2}\right]$ is the Kawski-Chamma-Viallete's polarity function.

In all above equations $\overline{\upsilon_a}$ and $\overline{\upsilon_f}$ are absorption and the fluorescence maxima wavenumbers (in cm^{-1}) respectively. n and ε are the refractive index and the dielectric constant of solvent respectively. From Eqs. (2), (3) and (4) graphs of $\left(\overline{\upsilon_a} - \overline{\upsilon_f}\right)$ versus $\phi\ (\varepsilon,\ n)$, $\left(\overline{\upsilon_a} - \overline{\upsilon_f}\right)$ versus $\phi_1\ (\varepsilon,\ n)$ and

Table 3 Spectral shift data for TMC molecule in different solvents

Solvent	$\overline{\upsilon_a}$ (cm^{-1})	$\overline{\upsilon_f}$ (cm^{-1})	$\overline{\upsilon_a} - \overline{\upsilon_f}$ (cm^{-1})	$\frac{\overline{\upsilon_a}+\overline{\upsilon_f}}{2}$ (cm^{-1})
Trichloroethylene	29411.76	23174.97	6237.70	26293.36
Benzene	29411.76	23185.71	6231.29	26298.73
Toluene	29325.51	23089.35	6236.141	26207.43
Dioxin	29411.76	23174.97	6237.79	26293.36
Tetrahydrofuran	29411.76	23169.60	6242.16	26290.68
Dimethyl sulphoxide	29154.51	22904.20	6250.31	26029.35
Acetone	29411.76	23158.86	6253.90	26285.31
Acetonitrile	29239.76	22988.50	6251.26	26114.13
Methanol	29411.76	23148.14	6253.10	26279.95

Table 4 Solvent parameters and calculated polarity functions

Solvent	Dielectric constant (ε)	Refractive index (n)	$\Phi(\varepsilon, n)$	$\Phi_1(\varepsilon, n)$	$\Phi_2(\varepsilon, n)$
Trichloroethylene	3.40	1.476	0.193	0.547	0.600
Benzene	2.30	1.501	0.032	0.006	0.341
Toluene	2.40	1.497	0.015	0.033	0.351
Dioxin	2.30	1.421	0.019	0.039	0.306
Tetrahydrofuran	7.50	1.404	0.209	0.547	0.548
Dimethyl sulphoxide	47.24	1.479	0.263	0.841	0.744
Acetone	21.00	1.360	0.284	0.839	0.641
Acetonitrile	36.64	1.344	0.304	0.861	0.664
Methanol	33.70	1.329	0.330	0.857	0.652

$\left(\frac{\bar{\upsilon}_a+\bar{\upsilon}_f}{2}\right)$ versus $\phi_2(\varepsilon, n)$ gives the linear graphs with slopes m, m_1 and m_2 respectively and are given by

$$m = \left[\frac{2\left(\mu_e - \mu_g\right)^2}{hca^3}\right] \tag{5}$$

$$m_1 = \left[\frac{2\left(\mu_e - \mu_g\right)^2}{hca^3}\right] \tag{6}$$

$$m_2 = \left[\frac{2\left(\mu_e^2 - \mu_g^2\right)}{hca^3}\right] \tag{7}$$

where

μ_g	Ground state dipole moment.
μ_e	Excited state dipole moment.
h	Planck's constant.
c	Velocity of the light in vacuum.
a	Onsager cavity radius of the solute molecule which can be calculated by adding an atomic volume of the constituting the molecules as suggested by Edward [38].

From Eqs. (6) and (7) the value of μ_g and μ_e are given by

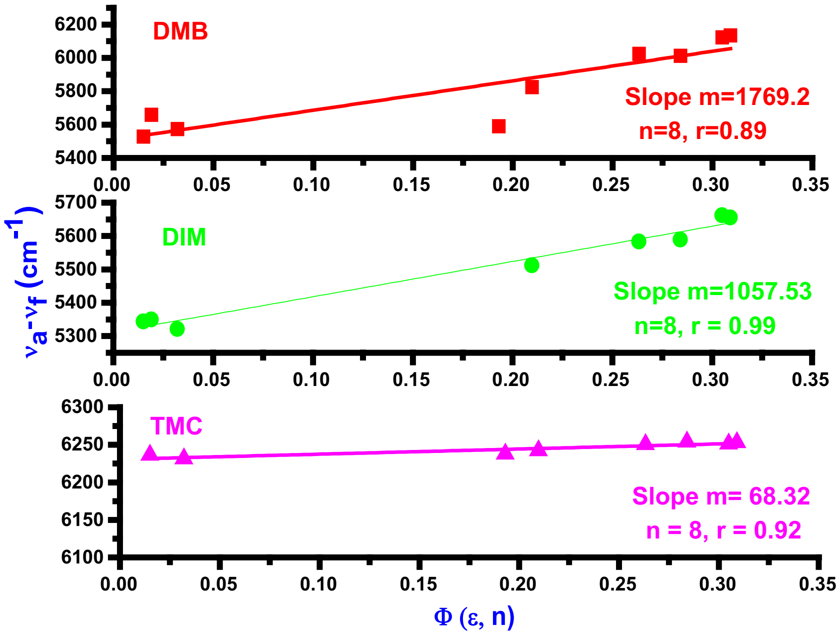
$$\mu_g = \frac{m_2 - m_1}{2}\left[\frac{hca^3}{2m_1}\right]^{\frac{1}{2}} \tag{8}$$

$$\mu_e = \frac{m_2 + m_1}{2}\left[\frac{hca^3}{2m_1}\right]^{\frac{1}{2}} \tag{9}$$

and

$$\frac{\mu_e}{\mu_g} = \left[\frac{m_2 + m_1}{m_2 - m_1}\right] \text{for } m_2 > m_1 \tag{10}$$

Fig. 2 Lippert's plots for all three molecules in different solvents



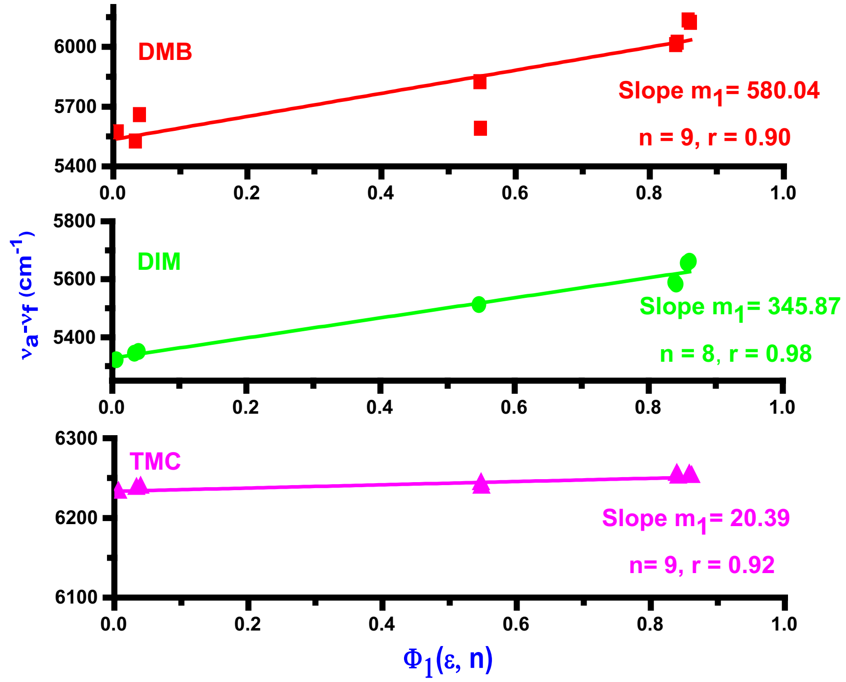


Fig. 3 Bakshiev's plots for all three molecules in different solvents

Guggenheim Method

The ground state dipole moment of all three solute molecules was calculated by Guggenheim method [39] which involves the refractive index and dielectric constant of the solute molecules and is given by

$$\mu_g^2 = \left[\frac{27kT}{4\pi N\left(\varepsilon_x+2\right)\left(n_x^2+2\right)}\right]\Delta \tag{11}$$

Where $\Delta = \left[\left(\frac{\varepsilon_{xy}-\varepsilon_x}{C}\right)_{C\to 0} - \left(\frac{n_{xy}^2-n_x^2}{C}\right)_{C\to 0}\right]$.

Here symbols k, T, N, ε, n and C are Boltzmann's constant, the temperature in Kelvin, Avogadro's number, dielectric constant, refractive index and concentration respectively. The suffixes x and xy refer to the solvent and solute respectively. Δ is extrapolated intercepts of the plots $\left(\frac{\varepsilon_{xy}-\varepsilon_x}{C}\right)$ versus C and $\left(\frac{n_{xy}^2-n_x^2}{C}\right)$ versus C with respect to infinite dilution (C→0).

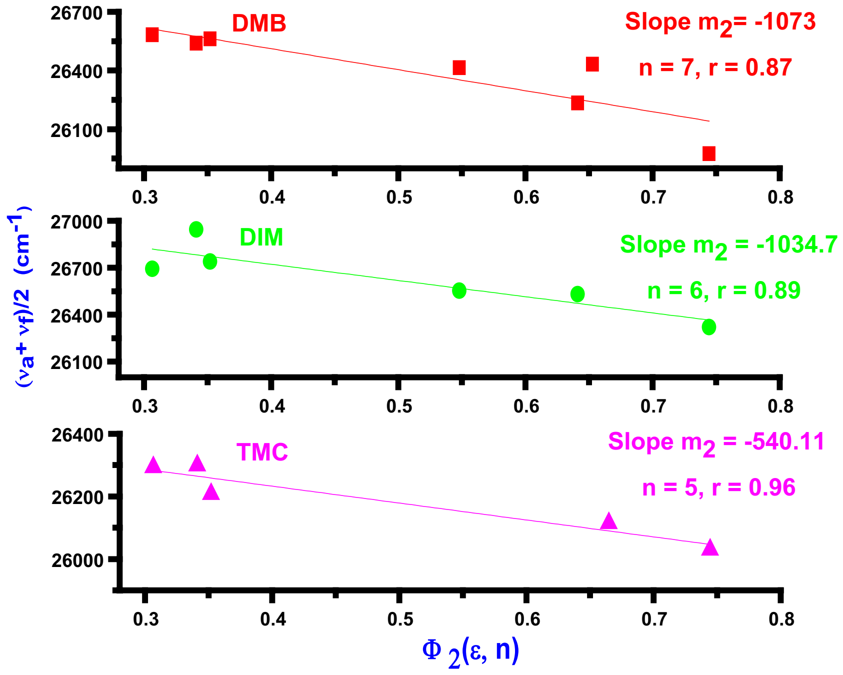


Fig. 4 KCV plots for all three molecules in different solvents

Table 5 Statistical treatment of the correlations for the solvent spectral shift of DMB, DIM and TMC molecules

Correlation	Slope (cm^{-1})	Intercept (cm^{-1})	Correlation coefficient (r)	Number of data (n)
Lippert's				
DMB	1769.2	5509	0.85	9
DIM	1057.5	53,123	0.99	8
TMC	68.32	6230.7	0.92	8
Bakhshiev's				
DMB	580.04	5534.8	0.89	9
DIM	345.87	5328.3	0.93	8
TMC	20.39	6233.4	0.92	9
KCV [a]				
DMB	−1073.0	26,941	0.87	7
DIM	−1034.7	27,136	0.89	6
TMC	−540.11	26,449	0.96	5

[a] Kawski-Chamma-Viallet correlation

Results and Discussion

The spectral shift data of absorption and emission spectra along with the Stokes shift of all three coumarin derivatives namely; 4-(2,6-Dibromo-4-methyl-phenoxymethyl)-benzo[h]chromen-2-one (DMB), 4-((3,4-dihydro-6,7-dimethoxyisoquinolin-1-yl)methyl)-6-methyl-2H-chromen-2-one (DIM) and 4-((p-tolyloxy)methyl)-6-methoxy-2H-chromen-2-one (TMC) are shown in Tables 1, 2, and 3 respectively. From these spectral shift data, one can identify the spectral transition namely; n→ π*, π→ π* etc [40, 41]. From Tables 1, 2, and 3 it can be notified that on the decrease in the solvent polarity which decreases the Stokes shift value of the solute molecules confirming that the π→π* transition in spectral levels. The shifting of the band occurs due to differences in the stabilization of ground and the excited state thus it causes a change in energy gap between these electronic states. And Table 4 summarizes the calculated values of the solvent polarity functions Φ (ε, n), Φ_1 (ε, n), Φ_2 (ε, n) along with the solvent parameters namely the dielectric constant (ε) and refractive index (n).

Figures 2, 3, and 4 shows the linear graphs of $\left(\overline{\upsilon_a} - \overline{\upsilon_f}\right)$ versus ϕ (ε, n), $\left(\overline{\upsilon_a} - \overline{\upsilon_f}\right)$ versus ϕ_1 (ε, n) and $\left(\frac{\overline{\upsilon_a}+\overline{\upsilon_f}}{2}\right)$ versus ϕ_2 (ε, n) for DMB, DIM and TMC molecules which gives the slopes m, m_1 and m_2 respectively. The slope, intercept, the number of data points and the statistical correlation values are shown in Table 5 and the good correlation value is obtained in all these cases, which clear that polarity functions exhibit quite good correlation with the spectral shifts for the selected number of data points. Generally, the deviation from the linearity may be due to specific solute–solvent interactions.

The ground state dipole moment value of all three solute molecules [DMB, DIM and TMC] obtained from the Guggenheim method were found to be 2.71, 2.33 and 2.47 D respectively and are shown in Table 6. The excited state dipole moment values were calculated using Eq. (9), Lippert Eq. (5), Bakshiev's Eq. (6) and Kawski-Chamma-Viallet's Eq. (7) are shown in Table 6. In all three solute molecules, the value of μ_e calculated from the Lippert's equation is larger, this may be due to the solute–solvent interaction evolved in the Lippert's equation [24]. The values of the ground and the excited state dipole moments of these three coumarin derivatives are different; this may be due to the structural difference between these three molecules. The change in dipole moment ($\Delta\mu$) from Eqs. (8) and (9) are also shown in Table 6. There is a large magnitude of the Stokes shift in the system studied, which may indicate that the excited state charge distribution is different from the ground state. In general, there will be an increase in Stokes

Table 6 The Onsager radius, the ground and excited state dipole moments of DMB, DIM and TMC molecules

Molecule	Radius(a) A°	μ_g^a (D)	μ_g^b (D)	μ_e^c (D)	μ_e^d (D)	μ_e^e (D)	μ_e^f (D)	Δμ^g (D)	h $\frac{\mu_e}{\mu_g}$
DMB	4.78	2.71	1.06	3.58	5.83	3.96	3.70	2.51	3.35
DIM	4.19	2.33	1.58	3.17	4.36	3.17	3.17	1.59	2.00
TMC	5.07	2.47	6.55	7.06	7.49	7.06	7.06	0.51	1.07

1 Debye = 3.33564×10^{-30}cm = 10^{-18} esu cm

[a] The ground state dipole moment calculated from Guggenheim method

[b] The ground state dipole moment calculated from the Eq. (8)

[c] The excited state dipole moment calculated from the Eq. (9)

[d] The experimental excited state dipole moment calculated from Lippert's Eq. (5)

[e] The experimental excited state dipole moment calculated from Bakshiev's Eq. (6)

[f] The experimental excited state dipole moment calculated from Kawski-Chamma Viallet's Eq. (7)

[g] The change in dipole moment calculated from the Eqs. (8) and (9)

[h] The ratio of the excited state and ground state dipole moment value calculated from the Eq. (10)

shift on increase in the solvent polarity, which shows that there is an increase in dipole moment on the excitation.

Conclusion

In summary, we have studied photophysical properties for novel coumarin derivatives namely; 4-(2,6-Dibromo-4-methyl-phenoxymethyl)-benzo[h]chromen-2-one(DMB), 4-((3,4-dihydro-6,7-dimethoxyisoquinolin-1-yl)methyl)-6-methyl-2H-chromen-2-one (DIM) and 4-((p-tolyloxy) methyl)-6-methoxy-2H-chromen-2-one (TMC). Absorption and fluorescence spectra of above said molecules were recorded in different polar and nonpolar solvents. From these spectral shift data, we observed that on the increase in solvent polarity the Stokes shift value also increases confirming spectral transition may be due to being $\pi\rightarrow\pi^*$ and we have calculated and compared the dipole moments of these three coumarin molecules in the ground and excited states by solvatochromic shift method and Guggenheim method. It is notified that all three Coumarin molecules possess higher dipole moment in the excited state than in the ground state. It is inferred that the excited state for above said molecules is more polar than the ground state. Further, an Eq. (10) can be used to estimate the value of excited state dipole moment by pre-knowledge of the value of ground state dipole moment, without a necessity of knowing the Onsager cavity radius of the solute molecule. The present investigations may shine in the design of nonlinear optical materials.

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