Synthesis and Conformation of Diethyl 2,3-diaryltetrahydro -4,5-isoxazoledicarboxylates

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The microwave assisted synthesis of diethyl 2,3-diaryltetrahydro-4,5-isoxazoledicarboxylates by the cycloaddition of diethyl maleate with appropriate nitrones was carried out. The conformational aspects of the title compounds were investigated by NMR and X-ray studies and the features are compared with a set of related tetrahydroisoxazoles highlighting the effect of substituents on the conformation of central five membered ring.

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Introduction.

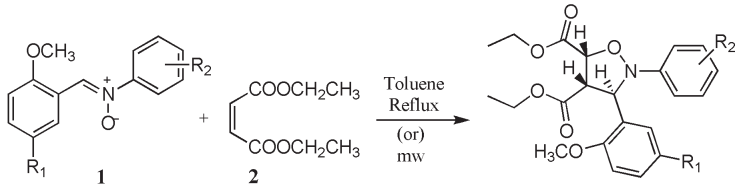
Five membered alicyclic compounds can have envelope or half chair conformation, the energy between them being very small. For the substituted systems also, there is no preference for any of these two arrangements. For example, 1,2-dichloropentane exists in equilibrium involving two envelope and two half chair arrangements [1]. Pseudo rotation leads to ten envelope forms and ten half chair forms. Thus there is no natural conformation of the five membered rings. The conformation of any one of the substituted cyclopentane or heterocyclopentane is likely to be different from that of any other. Isoxazolidine is no exception and no general preferred conformation is applicable to isoxazolidine, the substituents determining the preferred conformations in individual cases. A variety of examples have been cited in literature, in which the isoxazolidine ring has envelope [2-5] or twisted form [6]. The isoxazolidine ring has an envelope conformation when a fused lactone ring is six- or seven-membered, whereas an intermediate conformation between the envelope and the half-chair has been observed when the fused lactone is five-membered [7].

We have recently reported the synthesis of several isoxazolidines and investigated their regio and stereochemical features both by NMR and X-ray studies [8,9]. These investigated isoxazolidines have a methoxy group substituted in the *ortho* position of the 3-aryl ring and the substituents at positions 4 and 5 are *trans* to each other. In continuation of the above investigation, it has been proposed to prepare another new set of isoxazolidines in which *ortho* methoxy of 3-aryl system is retained but the stereochemistry of the substituents around 4 and 5 being changed to *cis*. With this aim, the 1,3-dipolar addition of diethyl maleate on appropriate nitrones, C-(5-substituted-2-methoxyphenyl)-N-aryl nitrones, have been carried out in the present investigation and the structural features of the resultant isoxazolidines have been analyzed and compared with the previously reported ones.

Results and Discussion.

The isoxazolidines, diethyl 2,3-diaryltetrahydro-4,5-isoxazoledicarboxylates (**3**) (Scheme 1), were obtained as the only product in appreciable yield (Table 1) from C-(5-substituted-2-methoxyphenyl)-N-aryl nitrones (**1**) and diethyl maleate (**2**) by heating the mixture in toluene. The

Scheme 1



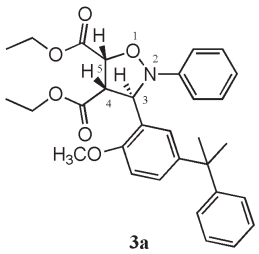
	R_1	R_2
a	$C(CH_3)_2Ph$	H
b	$C(CH_3)_2Ph$	*m*-CH_3
c	$C(CH_3)_2Ph$	*p*-CH_3
d	$C(CH_3)_3$	H
e	$C(CH_3)_2CH_2CH_3$	H

Table 1

Yields and Physical Constants of the Cycloadducts **3**

Compd	Microwave condition		Conventional method		
	Reaction time (min.)	Yield (%)	Reaction time (h)	Yield (%)	M.Pt. (ºC)
3a	5	94	7	85	96-97
3b	6	95	7	84	93-94
3c	6	93	7.5	86	115-16
3d	5	96	7.5	85	86-87
3e	5	95	7	84	Viscous liquid

reaction was also carried out under solvent free condition by mixing equimolar quantities of **1** and **2** using microwave irradiation (Table 1). The reaction gets completed in minutes with enhanced yield of the target compounds **3**. A multicomponent approach for the target molecule from the respective aldehyde, N-arylhydroxylamine and diethyl maleate has been attempted as observed in related isoxazolidines [9], but the reaction is not a clean one giving complex mixture. The isoxazolidines prepared have all been characterized by their IR and NMR spectra and complete assignment of all hydrogens and carbons have been achieved by 2D experiments. The NMR spectrum of **3a** (Figure 1) reveals the presence of two doublets, one at 4.88 ppm (J = 7.5 Hz) and another at 5.26 ppm (J = 5.1 Hz) along with a doublet of doublets at 3.76 ppm (J = 7.5, 5.1 Hz). The two doublets are identified as H-5 and H-3 respectively from HMBC experiments, as the latter makes strong contours with aromatic carbons. The coupling constant suggests that the stereochemistry of substituents around C-3 and C-4 is *trans*. The stereochemistry between C-4 and C-5 substituents can be expected to be *cis*, but the coupling constant is relatively smaller than anticipated. This can be accounted if the system assumes a preferred envelope arrangement keeping the C-5 carbon out of plane, in which case the dihedral angle between *cis* H-4 and H-5 could be around 60º accounting for the small coupling constant. This is also confirmed by X-ray analysis (*vide infra*). It is interesting to note that only one of the methylene of the carbethoxy groups that attached to the C-5 alone exhibits the diastereotopic nonequivalence, while the other appears as a neat quartet. The diastereotopic nonequivalence has also been noticed with the hydrogens and carbons of gem dimethyl group attached to the 3-aryl ring in **3a**, **3b**, **3c** and **3e**.



3a

Figure 1

It can be observed from the crystal X-ray analysis of **3a** (Figure 2) that the central five membered isoxazolidine ring has an envelope form with C-5 going out of plane [10]. Analysis of the crystal structures of isoxazolidines we have reported in which the C-3 aryl ring having a methoxy group at *ortho* position reveals interesting conformational difference in the central five membered ring. Table 2 summarizes the torsional angle between the ring atoms and the coupling constant for the H-3, H-4 and H-5 hydrogens of six different isoxazolidines **3a** [10], **4**, **5** [8], **6** [8], **7** [8] and **8** [9,11] (Figure 3). Though the NMR features of **4**, 3-[2-methoxy-5-(*t*-pentyl)phenyl]-4-nitro-2,5-diphenyltetrahydroisoxazole have already been discussed [8], the crystal features (Figure 4) are analyzed in the present investigation and the crystal data and structure refinement are presented in Table 3 [12]. In this case, the central five membered isoxazolidine ring has a half chair form rather than an envelope structure with the three carbon atoms in one plane and the nitrogen and oxygen atoms going out of plane in two different directions. The torsional angles between the planes N-C_3 & C_4-C_5 is 14.05˚ and that between C_3-C_4 & C_5-O is 9.93˚ (Table 2) clearly indicate that the nitrogen and oxygen atoms are out of plane with reference to the ring carbon atoms. The stereochemistry of the substituents around C_3, C_4 and C_5 are *trans* to each other and the alternative substituents are eclipsed to each other. All the three phenyl rings are almost perpendicular to each other.

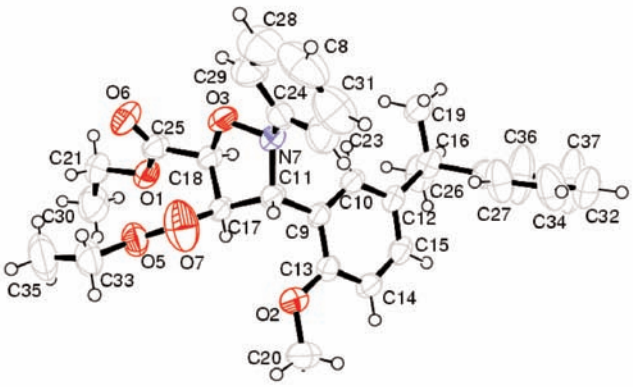


Figure 2. ORTEP Diagram of Diethyl 3-[2-methoxy-5-(1-methyl-1-phenylethyl)phenyl]-2-phenyltetrahydro-4,5-isoxazoledicarboxylate (**3a**).

It is to be noted that the analogous compound **5**, 3-[5-(*t*-butyl)-2-methoxyphenyl]-4-nitro-2,5-diphenyltetrahydroisoxazole has a similar half chair conformation but compound **6**, 3-[2-methoxy-5-(*t*-pentyl)phenyl]-5-(2-methoxy-

Table 2

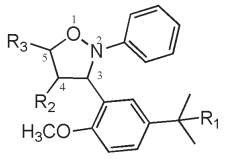
Selected Torsional Angles and Coupling Constants of Isoxazolidines **3a** to **8**

Compd	Dihedral angles (deg)					Coupling constants (Hz)		Ref.
	O-N & C_3-C_4	N-C_3 & C_4-C_5	C_3-C_4 & C_5-O	C_4-C_5 & O-N	C_5-O & N-C_3	$J_{3,4}$	$J_{4,5}$	
3a	1.27	23.53	-41.50	43.62	-28.35	5.1	7.5	[10]
4	-33.11	14.05	9.93	-31.47	41.38	5.4	9.0	[8]
5	-33.21	13.75	10.52	-32.43	42.05	2.4	6.3	[8]
6	-31.63	17.62	2.30	-22.55	34.70	2.7	6.3	[8]
7	22.24	2.59	-26.12	41.72	-40.61	2.7	5.7	[8]
8	-20.97	-2.50	24.76	-39.48	38.65	5.2	9.2	[11]

phenyl)-4-nitro-2-phenyltetrahydroisoxazole (Figure 3), has a perfect envelope structure. In the case of **6**, the carbon atoms C_3, C_4, C_5 and the oxygen atom lie in one plane leaving the nitrogen atom out of plane. The torsional angle between C_3-C_4 and C_5-O is only 2.30˚ indicating the envelope conformation for the heterocyclic ring in **6**. The observed conformational change could be due to the presence of the methoxy group at the *ortho* position of the 5-aryl ring. The selected torsional angles for **5** and **6** have been included in the Table 2.

It is interesting to note that compounds **7**, [3-[2-methoxy-5-(1-methyl-1-phenylethyl)phenyl]-5-(4-methylphenyl)-2-phenyltetrahydro-4-isoxazolyl](phenyl)methanone and **8**, 3-[2-methoxy-5-(1-methyl-1-phenylethyl)phenyl]-2,5-diphenyltetrahydro-4-isoxazolyl(2-thienyl)methanone both of them having aroyl group instead of nitro group at position 4, have entirely different conformation compared to **4**, **5** or **6** in their solid states. The torsional angle between N-C_3 and C_4-C_5 is 2.59˚ and -2.50˚ respectively for the compounds **7** and **8**. These minimum torsional angles can be attributed to an envelope conformation for **7** and **8** with oxygen atom being out of plane. The torsional angles around the isoxazolidine ring of compounds **7** and **8** have also been included in table 2.

The cycloadduct diethyl 3-[2-methoxy-5-(1-methyl-1-phenylethyl)phenyl]-2-phenyltetrahydro-4,5-isoxazole-



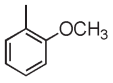
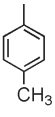
Compd	R_1	R_2	R_3
3a		$COOCH_2CH_3$	$COOCH_2CH_3$
4	CH_2CH_3	NO_2	
5	CH_3	NO_2	
6	CH_2CH_3	NO_2	
7			
8			

Figure 3

Table 3

Crystal Data and Structure Refinement of **4**

Empirical formula	$C_{27}H_{30}N_2O_4$	
Formula weight	446.53	
Temperature	295(2) K	
Wavelength	0.71073 A	
Crystal system	triclinic	
Space group	P-1	
Unit cell dimensions	a = 10.845(5) A	alpha = 112.27(3) deg.
	b = 11.405(4) A	beta = 109.63(4) deg.
	c = 11.715(5) A	gamma = 95.92(3) deg.
Volume	1218.0(9) A^3	
Z	2	
Density (calculated)	1.218 Mg/m^3	
Absorption coefficient	0.082 mm^-1	
F(000)	476	
Crystal size	0.53 x 0.43 x 0.33 mm	
Theta range for data collection	2.47 to 23.97 deg.	
Index ranges	0<=h<=12, -13<=k<=12, -13<=l<=12	
Reflections collected	4103	
Independent reflections	3796 [R(int) = 0.0161]	
Absorption correction	Psi-scans	
Max. and min. transmission	0.9990 and 0.9475	
Refinement method	Full-matrix least-squares on F^2	
Data / restraints / parameters	3796 / 4 / 323	
Goodness-of-fit on F^2	1.067	
Final R indices [I>2sigma(I)]	R1 = 0.0563, wR2 = 0.1481	
R indices (all data)	R1 = 0.0736, wR2 = 0.1629	
Extinction coefficient	0.025(4)	
Largest diff. peak and hole	0.310 and -0.212 e.A^-3	

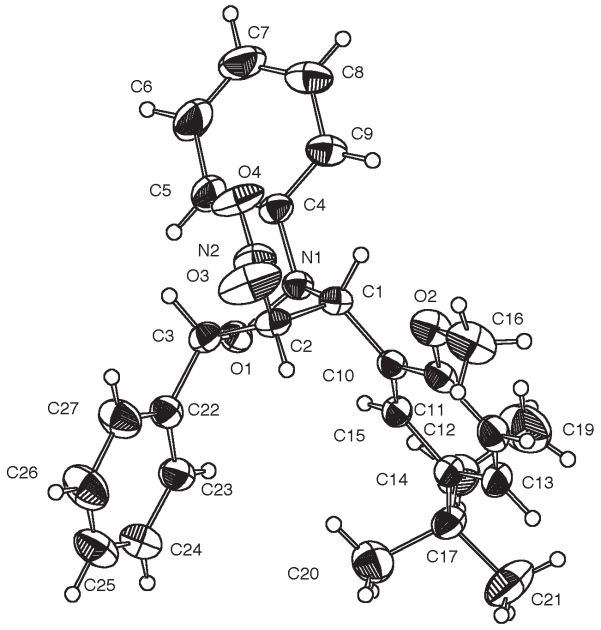
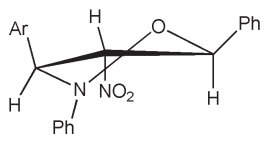


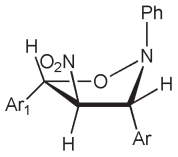
Figure 4. ORTEP diagram of 3-[2-methoxy-5-(*t*-pentyl)phenyl]-4-nitro-2,5-diphenyltetrahydroisoxazole (**4**).

dicarboxylate **3a** described in the present investigation has shown to have an entirely different conformational arrangement [10]. Though this compound also exists in an envelope form, it is the C_5 carbon and not oxygen or nitrogen that goes out of plane in this case. The torsional angle between O-N and C_3-C_4 is 1.27˚. In this arrangement the system avoids the possible eclipsed interaction between the *cis* carbethoxy groups.

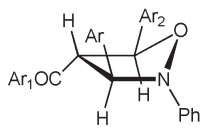
In conclusion, it is noticed that different 2,3,4,5-tetra-substituted isoxazolidines have different conformational arrangements (Figure 5) and the structure of the central five membered ring depends on the substituents present in the isoxazolidine ring. If the substituent at position 4 is nitro group, the compound prefers to exist in a half chair (**4** and **5**) or envelope with nitrogen atom being out of plane (**6**). On the other hand, if the C_4 substituent is aroyl, the isoxazolidines have an envelope conformation with oxygen atom going out of plane (**7** and **8**). In contrast to the above observations, if the substituents at C_4 and C_5 are *cis* carbethoxy groups, the molecule prefers an envelope structure with C_5 carbon left out of plane (**3a**).



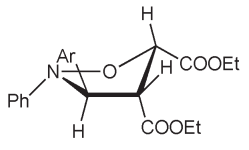
4 Ar = 2-Methoxy-5-*t*-pentylphenyl
5 Ar = 2-Methoxy-5-*t*-butylphenyl



6 Ar = 2-Methoxy-5-*t*-pentylphenyl
Ar_1 = 2-Methoxyphenyl



7 Ar = 2-Methoxy-5-cumylphenyl
Ar_1 = Phenyl, Ar_2 = 4-Methylphenyl
8 Ar = 2-Methoxy-5-cumylphenyl
Ar_1 = 2-Thienyl, Ar_2 = Phenyl



3a Ar = 2-Methoxy-5-cumylphenyl

Figure 5. Solid state conformational structures of isoxazolidines **3a** to **8**.

EXPERIMENTAL

Melting points are uncorrected. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker 300 MHz instrument in $CDCl_3$ using TMS as internal standard. Chemical shifts are given in parts per million (δ-scale) and coupling constants are given in Hertz. The single crystal X-ray data set was collected on a Nonius MACH3 Kappa diffractometer with Mo Kalpha radiation (λ = 0.71073 A). The structure was solved by direct methods using SHELXS-86 and refined by full matrix least squares on F^2 by SHELXL-93. The molecular views were realized by ZORTEP. IR Spectra were recorded on a Jasco FT-IR instrument (KBr pellets for solids and CCl_4 solution for viscous liquid). Elemental analyses were carried out on a Vario EL III instrument. Column chromatography was carried out in silica gel (60-120 mesh) using pet ether-ethyl acetate as eluent.

Cycloaddition of *C*-(5-substituted-2-methoxyphenyl)-*N*-aryl Nitrones (1) with Diethyl Maleate (**2**).

A. Conventional Method.

General Procedure: A mixture of 0.005 mole C-(5-substituted-2-methoxyphenyl)-N-aryl nitrone (**1**) and 0.005 mole of diethyl maleate (**2**) was refluxed in 50 mL of dry toluene for the time period specified in Table 1. After completion of the reaction, the solvent was removed under reduced pressure. The product (**3**) was recrystallised from petroleum ether-ethyl acetate mixture (**3a** to **3d**) or purified through column chromatography (**3e**).

B. Microwave Irradiation Method.

General Procedure: A mixture of 0.002 mole of C-(5-substituted-2-methoxyphenyl)-N-aryl nitrone (**1**) and 0.002 mole of diethyl maleate (**2**) was irradiated in the microwave oven with 60% power for the time period specified in Table 1. The progress of the reaction was monitored by tlc. The pure product (**3**) was obtained as described above.

Diethyl 3-[2-Methoxy-5-(1-methyl-1-phenylethyl)phenyl]-2-phenyltetrahydro-4,5-isoxazoledicarboxylate (**3a**).

This compound was obtained as colorless crystals (petroleum ether-ethyl acetate mixture), ir (potassium bromide): 2977, 2931, 1751, 1598, 1492, 1255, 1211 cm^{-1}; ^{1}H nmr: δ 1.13 (t, 3H, J = 7.2 Hz), 1.24 (t, 3H, J = 7.2 Hz), 1.63 (s, 3H), 1.64 (s, 3H), 3.76 (dd, J = 7.5, 5.1 Hz), 3.79 (s, 3H), 4.03 (q, 2H, J = 7.2 Hz), 4.19-4.23 (m, 2H), 4.88 (d, J = 7.5 Hz), 5.26 (d, 1H, J = 5.1 Hz), 6.78 (d, 1H, J = 8.7 Hz), 6.93 (tt, 1H, J = 7.2, 0.9 Hz), 7.02 (dd, 2H, J = 8.7, 0.9 Hz), 7.10 (dd, 1H, J = 8.7, 2.4 Hz), 7.12-7.26 (m, 7H), 7.51 (d, 1H, J = 2.4 Hz); ^{13}C nmr: δ 14.34, 14.40, 31.21, 31.30, 42.85, 55.88, 58.25, 61.62, 61.84, 68.58, 77.54, 110.61, 117.10, 123.02, 125.96, 126.90, 127.05, 127.12, 127.87, 128.37, 128.75,

143.52, 150.60, 151.11, 154.96, 168.69, 170.14.

Anal. Calcd. for $C_{31}H_{35}NO_6$: C, 71.93; H, 6.82; N, 2.71. Found: C, 71.87; H, 6.86; N, 2.68.

Diethyl 3-[2-Methoxy-5-(1-methyl-1-phenylethyl)phenyl]-2-(3-methylphenyl)tetrahydro-4,5-isoxazoledicarboxylate (**3b**).

This compound was obtained as colorless crystals (petroleum ether-ethyl acetate mixture), ir (potassium bromide): 2971, 2933, 1752, 1602, 1492, 1255, 1207 cm^{-1}; ^1H nmr: δ 1.13 (t, 3H, J = 6.9 Hz), 1.25 (t, 3H, J = 6.9 Hz), 1.63 (s, 3H), 1.64 (s, 3H), 2.25 (s, 3H), 3.76 (dd, 1H, J = 7.5, 4.8 Hz), 3.79 (s, 3H), 4.04 (q, 2H, J = 6.9 Hz), 4.19-4.23 (m, 2H), 4.88 (d, 1H, J = 7.5 Hz), 5.23 (d, 1H, J = 4.8 Hz), 6.74-6.80 (m, 3H), 6.91 (s, 1H), 7.01-7.25 (m, 7H), 7.51 (d, 1H, J = 1.8 Hz); ^{13}C nmr: δ 14.35, 14.42, 22.01, 31.23, 31.33, 42.85, 55.89, 58.27, 61.62, 61.86, 68.57, 77.60, 110.66, 114.26, 117.85, 123.93, 125.97, 126.99, 127.05, 127.12, 127.91, 128.38, 128.60, 138.47, 143.47, 150.53, 151.13, 154.99, 168.72, 170.25.

Anal. Calcd. for $C_{32}H_{37}NO_6$: C, 72.29; H, 7.01; N, 2.63. Found: C, 72.35; H, 7.10; N, 2.68.

Diethyl 3-[2-methoxy-5-(1-methyl-1-phenylethyl)phenyl]-2-(4-methylphenyl)tetrahydro-4,5-isoxazoledicarboxylate (**3c**).

This compound was obtained as colorless crystals (petroleum ether-ethyl acetate mixture), ir (potassium bromide): 2971, 2935, 1752, 1604, 1500, 1249, 1207 cm^{-1}; ^1H nmr: δ 1.15 (t, 3H, J = 6.9 Hz), 1.26 (t, 3H, J = 7.2 Hz), 1.63 (s, 6H), 2.24 (s, 3H), 3.75 (dd, 1H, J = 7.5, 5.4 Hz), 3.77 (s, 3H), 4.05 (q, 2H, J = 6.9 Hz), 4.21-4.25 (m, 2H), 4.89 (d, 1H, J = 7.5 Hz), 5.15 (d, 1H, J = 5.4 Hz), 6.77 (d, 1H, J = 8.4 Hz), 6.92-6.99 (m, 4H), 7.07-7.26 (m, 6H), 7.49 (d, 1H, J = 2.1 Hz); ^{13}C nmr: δ 14.38, 14.44, 21.14, 31.20, 31.29, 42.84, 55.86, 58.39, 61.56, 61.80, 68.92, 77.52, 110.56, 118.18, 125.93, 126.71, 127.06, 127.24, 127.72, 128.36, 129.32, 133.09, 143.42, 147.88, 151.13, 155.10, 168.73, 170.36.

Anal. Calcd. for $C_{32}H_{37}NO_6$: C, 72.29; H, 7.01; N, 2.63. Found: C, 72.25; H, 7.08; N, 2.66.

Diethyl 3-[5-(*t*-butyl)-2-methoxyphenyl]-2-phenyltetrahydro-4,5-isoxazoledicarboxylate (**3d**).

This compound was obtained as colorless crystals (petroleum ether-ethyl acetate mixture), ir (potassium bromide): 2964, 2908, 1749, 1594, 1496, 1257, 1207 cm^{-1}; ^1H nmr: δ 1.15 (t, 3H, J = 7.2 Hz), 1.25 (t, 3H, J = 7.2 Hz), 1.27 (s, 9H), 3.75 (dd, 1H, J = 7.5, 4.8 Hz), 3.82 (s, 3H), 4.02 (q, 2H, J = 7.2 Hz), 4.18-4.22 (m, 2H), 4.91 (d, 1H, J = 7.5 Hz), 5.35 (d, 1H, J = 4.8 Hz), 6.84 (d, 1H, J = 8.4 Hz), 6.93 (t, 1H, J = 7.2 Hz), 7.08 (d, 2H, J = 7.5 Hz), 7.19 (t, 2H, J = 7.5 Hz), 7.30 (dd, 1H, J = 8.4, 2.4 Hz), 7.63 (d, 1H, J = 2.4 Hz); ^{13}C nmr: δ 14.29, 14.38, 31.92, 34.65, 55.87, 58.30, 61.62, 61.83, 68.39, 77.69, 110.50, 116.61, 122.74, 125.62, 125.90, 127.19, 128.77, 144.04, 150.86, 154.68, 168.69, 170.07.

Anal. Calcd. for $C_{26}H_{33}NO_6$: C, 68.55; H, 7.30; N, 3.07. Found: C, 68.51; H, 7.28; N, 3.02.

Diethyl 3-[2-Methoxy-5-(*t*-pentyl)phenyl]-2-phenyltetrahydro-4,5-isoxazoledicarboxylate (**3e**).

This compound was obtained as a viscous liquid, ir (CCl_4): 2963, 2904, 1752, 1592, 1498, 1251, 1204 cm^{-1}; ^1H nmr: δ 0.63 (t, 3H, J = 7.5 Hz), 1.12 (t, 3H, J = 7.2 Hz), 1.23 (s, 6H), 1.25 (t, 3H, J = 7.2 Hz), 1.58 (q, 2H, J = 7.5 Hz), 3.76 (dd, 1H, J = 7.2, 4.8 Hz), 3.75 (s, 3H), 4.05 (q, 2H, J = 7.2 Hz), 4.19-4.23 (m, 2H), 4.92 (d, 1H, J = 7.2 Hz), 5.32 (d, 1H, J = 4.8 Hz), 6.84 (d, 1H, J = 8.4 Hz), 6.92 (t, 1H, J = 7.2 Hz), 7.07 (d, 2H, J = 8.4 Hz), 7.16-7.28 (m, 3H), 7.54 (d, 1H, J = 2.4 Hz); ^{13}C nmr: δ 9.51, 14.31, 14.40, 28.91, 29.03, 37.28, 37.83, 55.84, 58.28, 61.60, 61.82, 68.57, 77.65, 110.50, 116.78, 122.81, 126.31, 126.69, 126.93, 128.74, 142.27, 150.75, 154.67, 168.70, 170.11.

Anal. Calcd. for $C_{27}H_{35}NO_6$: C, 69.06; H, 7.51; N, 2.98. Found: C, 69.12; H, 7.48; N, 2.95.

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[12] Crystal data have been deposited to CCDC, number 236451.