Direct Coupling of Functionalized Organolithium Compounds with Aryl and Vinyl Halides

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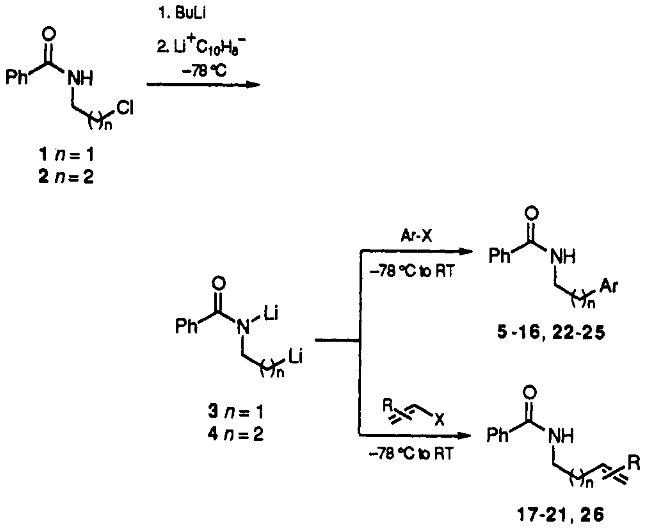
The reaction between β- and γ-nitrogen-functionalized and γ- and ϵ-oxygen-functionalized organolithium compounds **3**, **4**, **30–32** and different aromatic, heteroaromatic, and vinylic halides affords directly the corresponding substitution products: functionalized benzamides **5–26** and alcohols **33–37**. Symmetrical and mixed products of double coupling **38–40** were also prepared from 1,4-diiodobenzene. The formation of alkyl halides as intermediates has been verified. Aryl or vinyl halides giving rise to unstable aryl or vinyllithium reagents were unsuccessful in the coupling reaction.

Carbon–carbon bond formation by reaction of organometallic compounds with organic halides is a widely used method in organic synthesis.[1] The success of this reaction between a C(sp^3)-organometallic and a C(sp^2)-halide relies on the presence of catalytic amounts of a transition metal complex or salt. Copper,[2] palladium,[3] nickel,[4] and iron[5] catalysts are the most frequently employed in this cross-coupling reaction. However, some rare and isolated examples of the uncatalyzed direct coupling of alkylmetals with aryl and vinyl halides have been reported. The coupling reaction of pentylmagnesium bromide with β-bromostyrene occurred with moderate yield.[6] Methyllithium reacted more efficiently with iodobenzene and 1-iodonaphthalene,[7] butyllithium and ethyllithium gave the cross-coupled products with different aryl halides,[8] and the reaction of octyllithium with 1-bromocyclopentene led to 1-octylcyclopentene.[9] Presumably, these coupling reactions proceed through halogen-metal exchange followed by nucleophilic substitution. More recently,[10] the uncatalyzed cross-coupling of indium trialkyls with chloroalkenes has been reported and suggested to proceed *via* a multistep addition–elimination type pathway.

For several years we have been involved in the study of functionalized organolithium compounds, and we have described the preparation of different β-, γ-, δ-, and ϵ-oxygen- or nitrogen-functionalized systems and their reaction with electrophiles.[11] Continuing this study, we became interested in the reaction of these dianionic intermediates with aromatic and vinylic halides, which would broaden its synthetic applications. In this paper, we report a general study of this coupling reaction,[12] which proceeds directly without the assistance of transition metal catalysts.

Scheme I



Results and Discussion

As shown in the Scheme I, β- and γ-nitrogen-functionalized organolithium compounds **3** and **4**, respectively, were generated from the corresponding chloro benzamide **1** and **2** by successive treatment with butyllithium and lithium naphthalenide at -78 °C.[13,14] The *in situ* reaction of these dianions **3** and **4** with different aromatic and vinylic halides (-78 °C to room temperature) affords directly the corresponding coupling products **5–26** (Tables I and II). The coupling reaction works better with iodide and bromide derivatives than with chloride compounds (entries 1–3, Table I). With bromoiodobenzenes, selective coupling at the carbon–iodine bond occurs (entries 4, 10, Table I and entry 3, Table II). In the reaction of organolithium **4** with 1-bromo-4-iodobenzene (entry 3, Table II), a minor product

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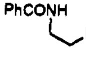
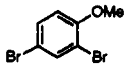
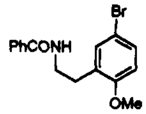
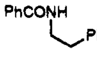
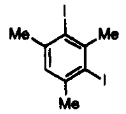
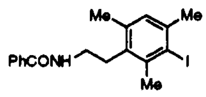
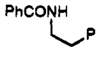
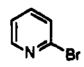
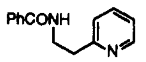
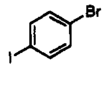
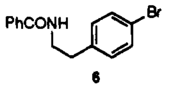
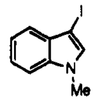
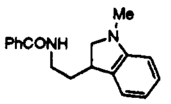
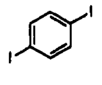
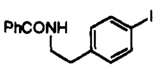
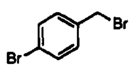
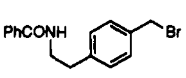
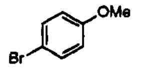
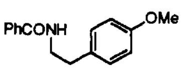
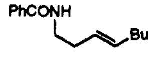
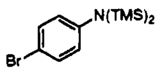
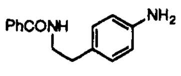
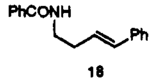
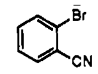
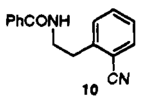
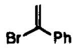
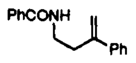
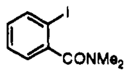
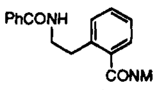
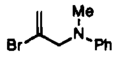
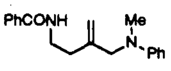
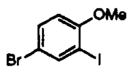
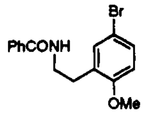
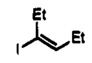
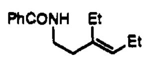
(14) The lithiation reaction can alternatively be carried out using a catalytic amount of naphthalene. Yus, M.; Ramón, D. J. *J. Chem. Soc., Chem. Commun.* **1991**, 398.

0022-3263/93/1958-5976$04.00/0

Table I. Coupling Reaction of Organolithium 3 with Aryl and Vinyl Halides

entry	aryl or vinyl halide	products	yield[a,b] (%)	entry	aryl or vinyl halide	products	yield[a,b] (%)
1	PhI	 5	95 (75)	11		 12	85 (55)
2	PhBr	 5	95 (75)	12		 13	81 (66)
3	PhCl	 5	23[c]	13		 14	85 (58)
4		 6	88 (74)	14		 15	85 (69)
5		 7	95 (56)	15		 16	60 (41)
6		 8	91 (65)	16		 17	95 (79)
7		 9	*d* (53)[e]	17		 18	70 (55)
8		 10	*f* (40)	18		 19	98 (75)
9		 11	*f* (35)	19		 20	95 (78)
10		 12	70 (58)	20		 21	95 (73)

[a] Isolated yields based on starting material 1. [b] In parentheses are yields after chromatographic purification. [c] *N*-Ethylbenzamide (70%) was also formed. [d] Not calculated; the product obtained after usual workup was the *bis*-trimethylsilylated amine derivative of 9. [e] Isolated by acid–base extraction (1 N HCl, 1 N NaOH). [f] Not calculated; the reaction crude product was not clean.

24 resulting from the coupling at the carbon–bromine bond was formed in 17% yield. Different substituted aryl bromides and iodides also coupled smoothly in good yields (entries 5–10, Table I and entry 4, Table II); the moderate yields of compounds 10 and 11 could account for the higher reactivity toward organolithium reagents of the nitrile and amide groups. The coupling reaction with 2,4-dibromoanisole (entry 11) took place regioselectively at the *ortho* position, probably due to a complex-induced proximity effect (CIPE)[15] of the methoxy group. Sterically crowded iodide (entry 12) and heteroaryl halides (entries 13, 14) underwent clean coupling, too. Unexpectedly, 4-bromobenzyl bromide selectively reacted with organolithium 3 through the arylic bromide in the presence of the reactive benzylic bromide (entry 15). The couplings with vinyl bromides and iodides (entries 16–20, Table I and entry 5, Table II) are straightforward and proceed with retention of configuration at the double bond.

γ-Oxygen-functionalized[16] organolithium compounds 30 and 31 and ϵ-oxygen-functionalized[17] derivative 32 were analogously prepared from the corresponding chlorohydrins 27–29 by successive reaction with butyllithium and lithium naphthalenide at −78 °C. Treatment of these dianions with aryl or heteroaryl halides (−78 °C to room temperature) leads directly to the corresponding coupling products 33–37 (Scheme II and Table II). Disappointingly, the reaction of these oxygenated dianions with vinyl halides failed to produce any coupled product.[18]

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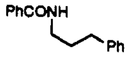
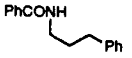
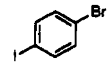
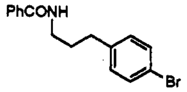
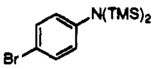
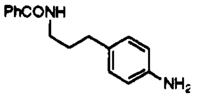
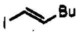
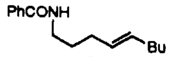
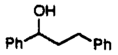
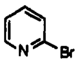
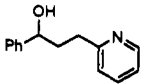
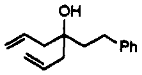
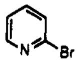
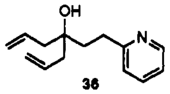
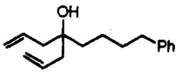
(16) (a) Barluenga, J.; Flórez, J.; Yus, M. *Synthesis* **1983**, 378. (b) Nájera, C.; Yus, M.; Seebach, D. *Helv. Chim. Acta* **1984**, *67*, 289. (c) Mudryk, B.; Cohen, T. *J. Org. Chem.* **1989**, *54*, 5657.

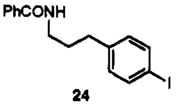
(17) Barluenga, J.; Rubiera, C.; Fernández, J. R.; Flórez, J.; Yus, M. *Synthesis* **1987**, 819.

(18) The cross-coupling products with vinyl halides have been obtained in the presence of Pd(PPh$_3$)$_4$ as catalyst. Unpublished results from our laboratory.

Table II. Coupling Reaction of Organolithiums 4, 30, 31, and 32 with Aryl and Vinyl Halides

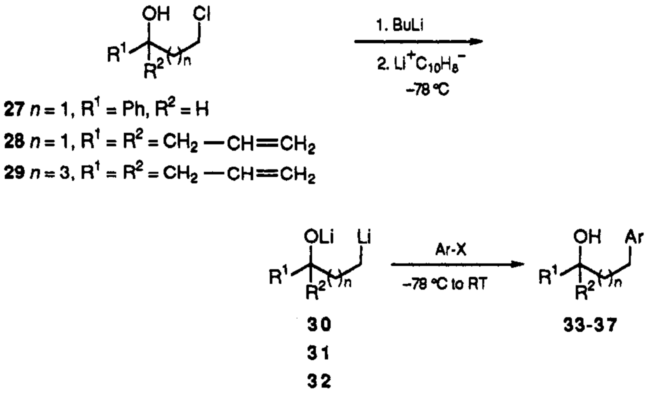
entry	organolithium compound	aryl or vinyl halide	products	yield^{a,b} (%)
1	**4**	PhI	 **22**	90 (70)
2	**4**	PhBr	 **22**	90 (70)
3	**4**		 **23**	68 (56)^{c}
4	**4**		 **25**	75 (67)
5	**4**		 **26**	82 (71)
6	**30**	PhI	 **33**	95 (74)
7	**30**		 **34**	95 (64)
8	**31**	PhI	 **35**	95 (70)
9	**31**		 **36**	92 (62)
10	**32**	PhI	 **37**	95 (65)

[a] Isolated yields based on the corresponding starting material 2, 27, 28, or 29. [b] In parentheses are yields after chromatographic purification. [c] Calculated yields. Product contaminated with 17% (14% after chromatographic purification) of iodine compound **24**; these products could not be separated by flash column chromatography or by recrystallization.

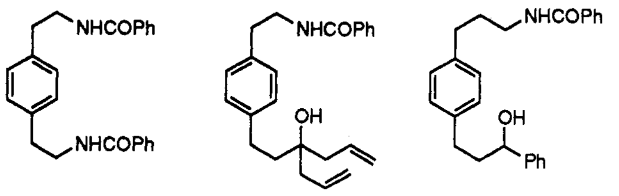


24

Scheme II



Scheme III



38 **39** **40**

Products of double coupling were obtained from 1,4-diiodobenzene and two equal or different functionalized organolithium compounds. Thus, the reaction of dianion **3** with 1,4-diiodobenzene (2:1 molar ratio) led to the symmetrical compound **38** [90% (80%)[19]]. Access to mixed coupling products is illustrated in the successive *in situ* reaction of compounds **31** or **30** with 1,4-diiodobenzene (1:1 molar ratio) and then with dianions **3** or **4**, respectively; by this method, products **39** (54%)[19] and **40** [65% (48%)[19]] were isolated (Scheme III).

This direct coupling reaction seems to proceed by a halogen–metal exchange (HME) pathway followed by nucleophilic substitution. In order to find some evidence for this mechanism we attempted to isolate the function-

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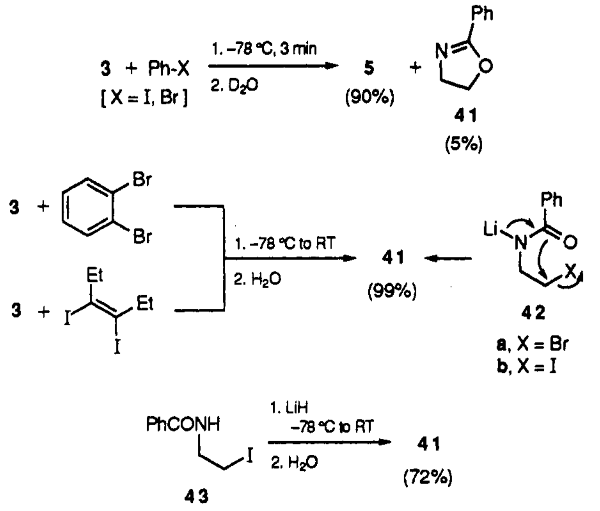
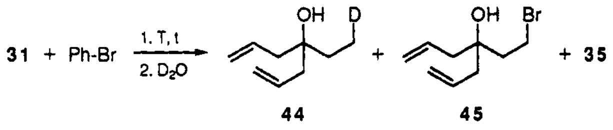
(19) See footnote b in Tables I and II.

Scheme IV



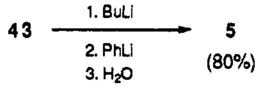
Scheme V



T (°C)	t (min)	44 : 45 : 35 ratio
-78	3	52 : 18 : 30
-78	60	34 : 16 : 50
-50	60	30 : 10 : 60

alized alkyl halide generated in the HME reaction before it could react with the simultaneously formed aryl or vinyllithium compound. We first studied the reaction of organolithium **3** with PhI and analyzed an aliquot 3 min after the addition of the iodide by quenching with deuterium oxide. Surprisingly, the coupling reaction was almost complete, yielding compound **5** (90%) and a small amount of product **41** (5%). A similar result was found in the reaction of **3** with PhBr (Scheme IV). And a very fast coupling occurred also in the reaction of dianion **4** with PhBr under the same experimental conditions (-78 °C, 3 min; 95% of **22**). Compound **41** could be easily identified by comparison with a sample which had been previously obtained, in almost quantitative yield, in the reaction of dianion **3** with 1,2-dibromobenzene or (*Z*)-3,4-diiodo-3-hexene (Scheme IV). At the same time, this outcome reveals that this coupling reaction is limited by the stability of the aryl or vinyllithium. Presumably, 2-oxazoline **41** is formed from intermediate **42** through intramolecular elimination of lithium halide. This was proved in an independent experiment in which iodo amide **43** was treated with lithium hydride between -78 °C and room temperature (Scheme IV). This result indirectly shows that functionalized alkyl halides are intermediates in these coupling reactions. Further direct evidence for this mechanism was observed with oxygen-functionalized organolithium compounds which have turned out to be less reactive than the nitrogen-functionalized ones in this coupling reaction. The results obtained with organolithium **31** and PhBr at different times and temperatures are indicated in Scheme V (the ratio of products was determined by capillary gas chromatography). Finally, evidence for the nucleophilic substitution step was gained in the reaction indicated in Scheme VI, which reproduces the reaction conditions after the HME step.

Scheme VI



In conclusion, we have demonstrated that different *N*-substituted benzamides and different substituted alcohols can be efficiently prepared by the uncatalyzed one-pot coupling reaction of functionalized organolithium compounds with aryl, heteroaryl, or vinyl halides. Different functional groups on the halide partner are tolerated, but substrates giving rise to unstable aryl or vinyllithiums in the HME step were unsuccessful. The formation of alkyl halides as intermediates in the coupling reaction has been confirmed.

Experimental Section

General. All reactions that involved organolithium derivatives were performed under a positive pressure of dry N_2 using oven-dried glassware. All liquid solutions and anhydrous solvents were transferred by oven-dried syringes. All solvents were freshly distilled from the appropriate drying agent before use.[20] THF was distilled from sodium benzophenone ketyl under N_2. Compound **1**, most of the aryl and vinyl halides, and other reagents were commercially available (Aldrich) of the best grade and were used without further purification. Starting materials **2**,[13] **27**,[21] **28**,[16a] and **29**,[17] lithium naphthalenide,[22] 4-bromo-*N*,*N*-bis(trimethylsilyl)aniline,[23] 4-bromo-2-iodoanisole,[24] 2,4-diiodo-1,3,5-trimethylbenzene,[24] 3-iodo-1-methylindole,[24] (*E*)-1-iodo-1-hexene,[25] (*E*)-3-iodo-3-hexene,[25] *N*-methyl-*N*-phenyl-2-bromo-2-propenylamine,[26] and (*Z*)-3,4-diiodo-3-hexene[27] were synthesized according to the literature methods. 2-Iodo-*N*,*N*-dimethylbenzamide was prepared from 2-iodobenzoyl chloride and an aqueous solution of dimethylamine in Et_2O. Drying of all organic extracts was carried out with anhydrous Na_2SO_4. The term in vacuo refers to solvent removal in a rotatory evaporator.

TLC was performed on aluminum-backed plates coated with silica gel 60 F_{254} (Merck or Scharlau). Components were located by observation of the plates under UV light and/or by treating the plates with I_2. Flash column chromatography was performed by using silica gel 60, 230–400 mesh (Merck or Scharlau) as described by Still.[28] GC analyses were carried out on a capillary column HP-ULTRA 2 (25 m, 0.2 mm i.e.; 0.33 μm film thickness). Melting points were obtained using open capillary tubes and are uncorrected. Unless otherwise stated, NMR spectra were recorded for $CDCl_3$ solutions at 200 or 300 MHz and ^{13}C NMR at 50.5 or 75.5 MHz, using tetramethylsilane (0.0 ppm, ^1H NMR) or chloroform-*d* (77.0 ppm, ^{13}C NMR) as internal standard; δ values are given in ppm and the coupling constants are recorded in hertz. Carbon multiplicities were obtained from DEPT experiments. Low-resolution electron impact mass spectrum data (EI-LRMS) were obtained at 70 eV and are reported as percent relative intensity to the base peak after the corresponding *m/e* value.

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General Procedure for the Coupling Reaction of Organolithium Compounds 3, 4, and 30–32 with Arylic and Vinylic Halides. To a solution of the corresponding chloro benzamide 1 and 2 or chlorohydrin 27–29 (2 mmol) in THF (30 mL) was added a solution of butyllithium in hexane (2.2 mmol) at −78 °C under N_2, and the mixture was stirred for 30 min at the same temperature. Then, a solution of lithium naphthalenide (4 mmol) in THF was added and stirring was continued for 6–8 h at −78 °C. To the resulting mixture was added a solution of the corresponding aryl or vinyl halide (2 mmol) in THF (20 mL) and it was stirred at −78 °C for 1 h and then allowed to rise to room temperature overnight. The reaction mixture was hydrolyzed and neutralized with 1 N HCl and extracted with ether. The organic layer was dried and concentrated in vacuo, and the residue was purified by flash column chromatography with a mixture of CH_2Cl_2/Et_2O, 2:1 as eluant. This method was used to prepare compounds **5–26** and **33–37** listed in Tables I and II. Yields are reported therein. Physical and spectral data for these compounds are provided as supplementary material.

In some cases (compounds **5**, **8**, **9**, **18**, and **19**), this procedure has been performed on a scale up to a 2.5 times larger with identical results.

General Procedure for the Double Coupling Reaction of Organolithium Compounds 3, 4, 30, and 31 with 1,4-Diiodobenzene. To a solution of the corresponding organolithium compound **3**, **31**, or **30** (2 mmol), prepared as described in the preceding general procedure, was added at −78 °C a solution of 1,4-diiodobenzene (2 mmol) in THF (20 mL) and the mixture was stirred at −78 °C for 1 h and then allowed to rise to room temperature overnight. Then, this reaction mixture was respectively added at −78 °C over a solution of the corresponding organolithium compound **3** or **4** (2 mmol) previously prepared as described above. The resulting mixture was stirred at −78 °C for 1 h and then warmed to 20 °C for 6–8 h. The reaction mixture was worked up as previously described. Yields are reported in the text. This method was used to prepare the following compounds:

1,4-Bis(2-benzamidoethyl)benzene (38): R_f 0.41 (AcOEt); mp 221–223 °C; ^{1}H NMR δ 2.9 (t, J = 7.0, 4H), 3.7 (q, J = 7.0, 4H), 6.2 (br s, 2H), 7.2–7.8 (3 m, 14H); ^{13}C NMR δ 35.3, 41.1, 126.7, 128.5, 129.1, 131.4, 134.5, 137.1, 167.4; MS *m/e* 372 (M^+, <1), 105 (100), 77 (70).

1-(3-Allyl-3-hydroxy-5-hexenyl)-4-(2-benzamidoethyl)benzene (39): R_f 0.43 (AcOEt); mp 70–72 °C; ^{1}H NMR δ 1.7 (m, 2H), 2.2 (s, 1H), 2.3 (d, J = 7.4, 4H), 2.7 (m, 2H), 2.85 (t, J = 6.9, 2H), 3.65 (q, J = 6.9, 2H), 5.1–5.2 (m, 4H), 5.8–6.0 (m, 2H), 6.85 (br s, 1H), 7.1–7.85 (3 m, 9H); ^{13}C NMR δ 29.1, 35.0, 40.9, 41.1, 43.4, 73.0, 118.4, 126.7, 128.2, 128.3, 128.6, 131.1, 133.4, 134.3, 136.0, 140.4, 167.4; MS *m/e* 322 (49), 159 (23), 117 (53), 105 (100).

1-(3-Benzamidopropyl)-4-(3-hydroxy-3-phenylpropyl)benzene (40): R_f 0.52 (CH_2Cl_2/Et_2O, 2:1); ^{1}H NMR δ 1.75–2.15 (m, 4H), 2.4–2.7 (m, 4H), 3.1–3.4 (m, 3H), 4.6 (m, 1H), 6.95–7.7 (3 m, 15H); ^{13}C NMR δ 30.6, 31.2, 32.6, 39.4, 40.3, 73.1, 125.5, 126.6, 126.8, 127.8, 127.9, 128.0, 130.8, 134.0, 138.4, 139.1, 144.5, 167.4; MS *m/e* 373 (M^+, <1), 251 (11), 130 (71), 105 (100).

Synthesis of *N*-(2-Iodoethyl)benzamide (43).[29] A mixture of *N*-(2-chloroethyl)benzamide (**1**, 2.65 g, 14.4 mmol), NaI (0.43 g, 2.8 mmol), and CH_3I (9 mL, 144 mmol) in *N*-methyl-2-pyrrolidinone (20 mL) was stirred at 70 °C for 96 h. The mixture was washed with aqueous $Na_2S_2O_3$ and extracted with CH_2Cl_2. The extracts were washed with brine, dried, and concentrated in vacuo. The residue was dried under reduced pressure to give pure product **43** (2.37 g, 61%) as a solid which was recrystallized from $EtOH-H_2O$: mp 104–106 °C; ^{1}H NMR δ 3.3 (t, J = 6.2, 2H), 3.7 (q, J = 6.3, 2H), 6.8 (br s, 1H), 7.25–7.8 (2 m, 5H); ^{13}C NMR δ 5.4, 42.0, 126.9, 128.5, 131.6, 133.9, 167.5; MS *m/e* 275 (M^+, 1), 149 (14), 105 (100), 77 (76).

Preparation of 2-Phenyl-4,5-dihydrooxazole (41) from 43. To a solution of *N*-(2-iodoethyl)benzamide (**43**, 0.37 g, 1.35 mmol) in THF (30 mL) at −78 °C was added LiH (0.011 g, 1.5 mmol) and the mixture was stirred for 10 h between −78 °C and room temperature. The reaction mixture was quenched with water and extracted with ether. The organic layer was dried and concentrated in vacuo and the residue was purified by flash column chromatography (CH_2Cl_2/Et_2O, 2:1) to afford **41** (0.14 g, 72%): R_f 0.46; ^{1}H NMR δ 3.8 (t, J = 7.0, 2H), 4.2 (t, J = 7.0, 2H), 7.1–7.9 (2 m, 5H); ^{13}C NMR δ 54.4, 67.2, 126.2, 128.0, 128.3, 131.5, 167.3; MS *m/e* 147 (M^+, 58), 117 (100), 105 (11), 77 (32).

Synthesis of *N*-(2-Phenylethyl)benzamide (5) from 43. To a solution of *N*-(2-iodoethyl)benzamide (**43**, 0.82 g, 3 mmol) in THF (20 mL) under N_2 at −78 °C was added a solution of butyllithium in hexane (1.2 mL, 3.3 mmol). The mixture was stirred for 30 min at this temperature and then a solution of phenyllithium in THF (20 mL, 3 mmol) was added. The reaction mixture was stirred overnight allowing the temperature to rise slowly to 20 °C. The resulting solution was quenched with a 1 N HCl solution and extracted with ether. The organic layer was dried and concentrated in vacuo, and the residue was dried under reduced pressure affording essentially pure product **5** (0.53 g, 80%).[30]

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Supplementary Material Available: Physical and spectral data for compounds **5–23**, **25**, **26**, and **33–37**; and copies of ^{1}H and ^{13}C NMR spectra of all compounds reported (36 pages). This material is contained in libraries on microfiche, immediately follows this article in the microfilm version of the journal, and can be ordered from the ACS; see any current masthead page for ordering information.

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[30] The level of purity of compounds is indicated by the inclusion of copies of NMR spectra presented in the supplementary material.