Supplementary Information:

Table S1. Average BP86 Calculated g-tensor and Cu(II) A-tensor Values of Model Complexes.[a,b,c]

		imidazole		1-methylimidazole		4-methylimidazole		
		gas	COSMO	gas	COSMO	gas	COSMO	Exp.[d]
g_z		2.093[1]	2.094[1]	2.093[1]	2.093[1]	2.095[2]	2.095[2]	2.215
	$+H_2O$	2.093[1]	2.093[1]	2.093[1]	2.093[1]	2.095[2]	2.094[2]	
g_y		2.029[1]	2.029[1]	2.029[1]	2.029[1]	2.029[2]	2.029[1]	2.050
	$+H_2O$	2.029[1]	2.029[1]	2.029[1]	2.029[1]	2.029[2]	2.029[1]	
g_x		2.025[1]	2.026[0.5]	2.025[1]	2.026[0.5]	2.026[1]	2.026[1]	2.050
	$+H_2O$	2.025[1]	2.026[0.5]	2.025[1]	2.026[1]	2.025[1]	2.026[1]	
A_z		-498[4]	-509[4]	-493[5]	-508[4]	-488[12]	-500[11]	(-)580
	$+H_2O$	-489[4]	-506[4]	-485[4]	-506[4]	-478[11]	-497[11]	
A_y		-30[5]	-35[4]	-25[6]	-34[4]	-26[3]	-34[3]	(-)70
	$+H_2O$	-16[14]	-32[4]	-10[18]	-32[4]	-8[18]	-30[3]	
A_x		-6[5]	-15[3]	1[5]	-13[3]	-2[9]	-13[8]	(-)70
	$+H_2O$	4[7]	-9[3]	2[9]	-9[3]	2[9]	-8[7]	

[a] A-tensor values in MHz

[b] Numbers in brackets are the standard deviation for each value.

[c] Standard deviations of g-tensor $x10^{-3}$.

[d] Experimental data from Ref. [1]

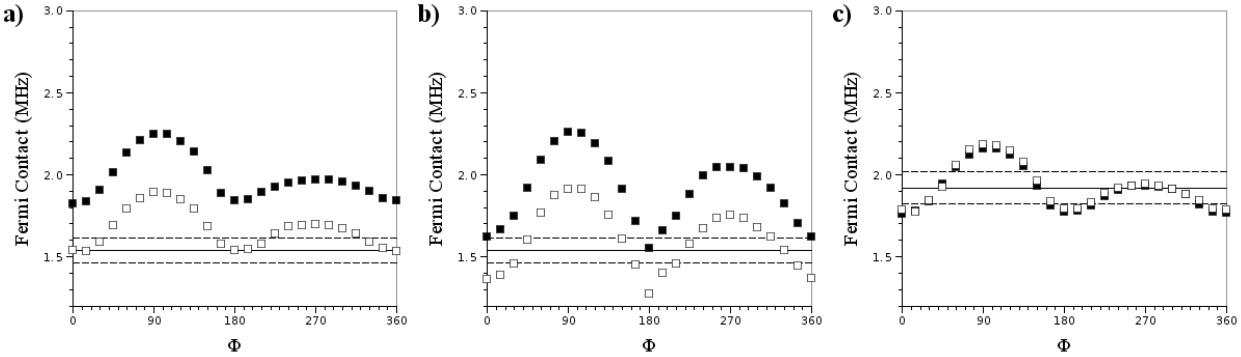

Figure S1. B3LYP calculated Fermi contact, A_{iso}, terms for the remote nitrogen atom, utilizing COSMO, of the a) imidazole b) 4-methylimidazole and c) 1-methylimidazole ligands. The closed, black, symbols are the calculations performed in the absence of a water molecule, while the open symbols are for calculations done with an explicit water molecule in close proximity to the remote nitrogen atom. The solid lines are experimental values from Ref. 1. Dashed lines represent a ±5% error in the experimentally determined values.

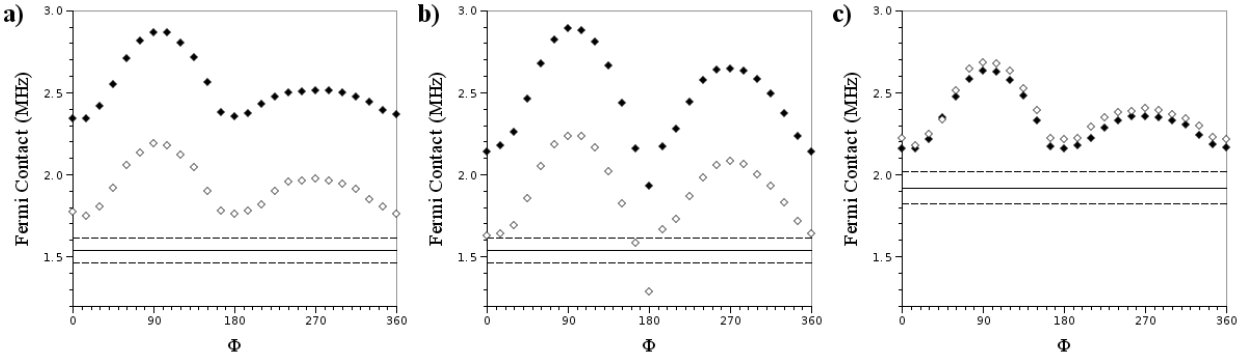

Figure S2. BP calculated Fermi contact, A_{iso}, terms for the remote nitrogen atom of the a) imidazole b) 4-methylimidazole and c) 1-methylimidazole ligands. The closed, black, symbols are the calculations performed in the absence of a water molecule, while the open symbols are for calculations done with an explicit water molecule in close proximity to the remote nitrogen atom. The solid lines are experimental values from Ref. 1. Dashed lines represent a ±5% error in the experimentally determined values.

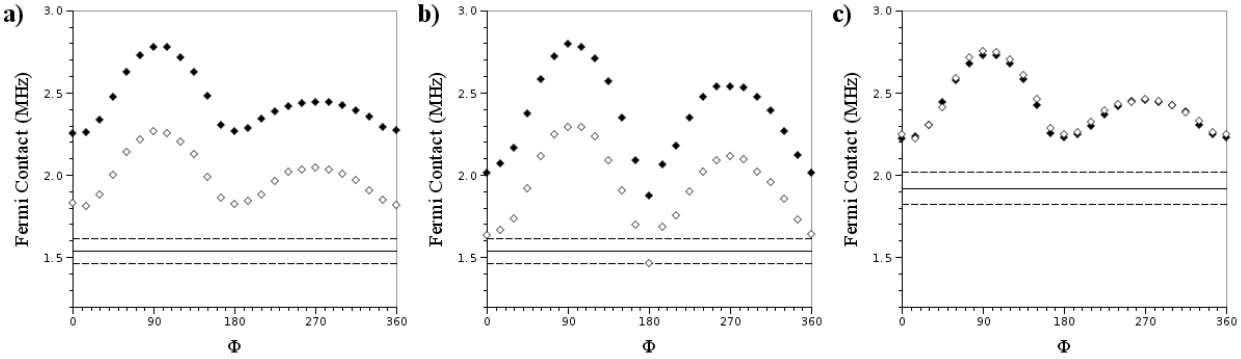

Figure S3. BP calculated Fermi contact, A_{iso}, terms for the remote nitrogen atom, utilizing COSMO, of the a) imidazole b) 4-methylimidazole and c) 1-methylimidazole ligands. The closed, black, symbols are the calculations performed in the absence of a water molecule, while the open symbols are for calculations done with an explicit water molecule in close proximity to the remote nitrogen atom. The solid lines are experimental values from Ref. 1. Dashed lines represent a ±5% error in the experimentally determined values.

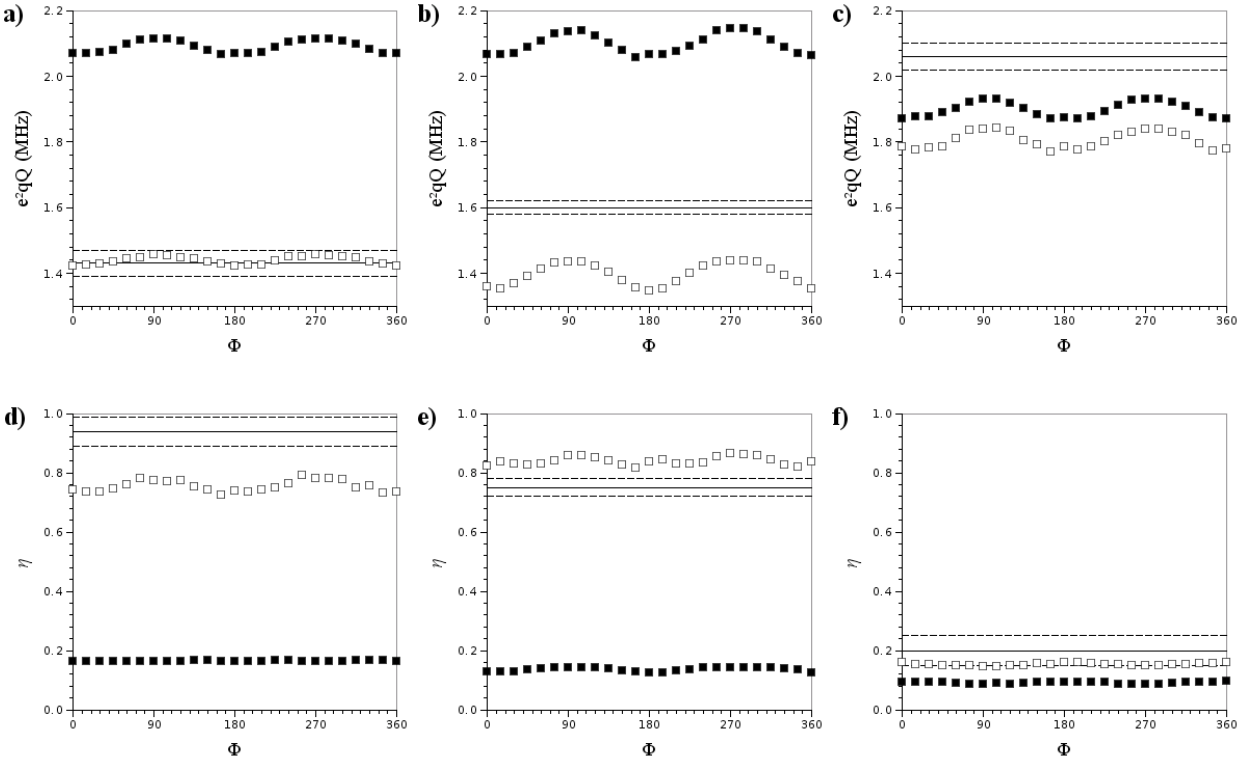

Figure S4. B3LYP calculated NQI, a) – c), and η, d) – f), for the remote nitrogen of the imidazole, 4-methylimidazole and 1-methylimidazole ligands, respectively. The closed, black, symbols are the calculations performed in the absence of a water molecule, while the open symbols are for calculations done with an explicit water molecule in close proximity to the remote nitrogen atom. The solid lines are experimental values and the dashed lines represent the experimentally determined error for each parameter from Ref. 1.

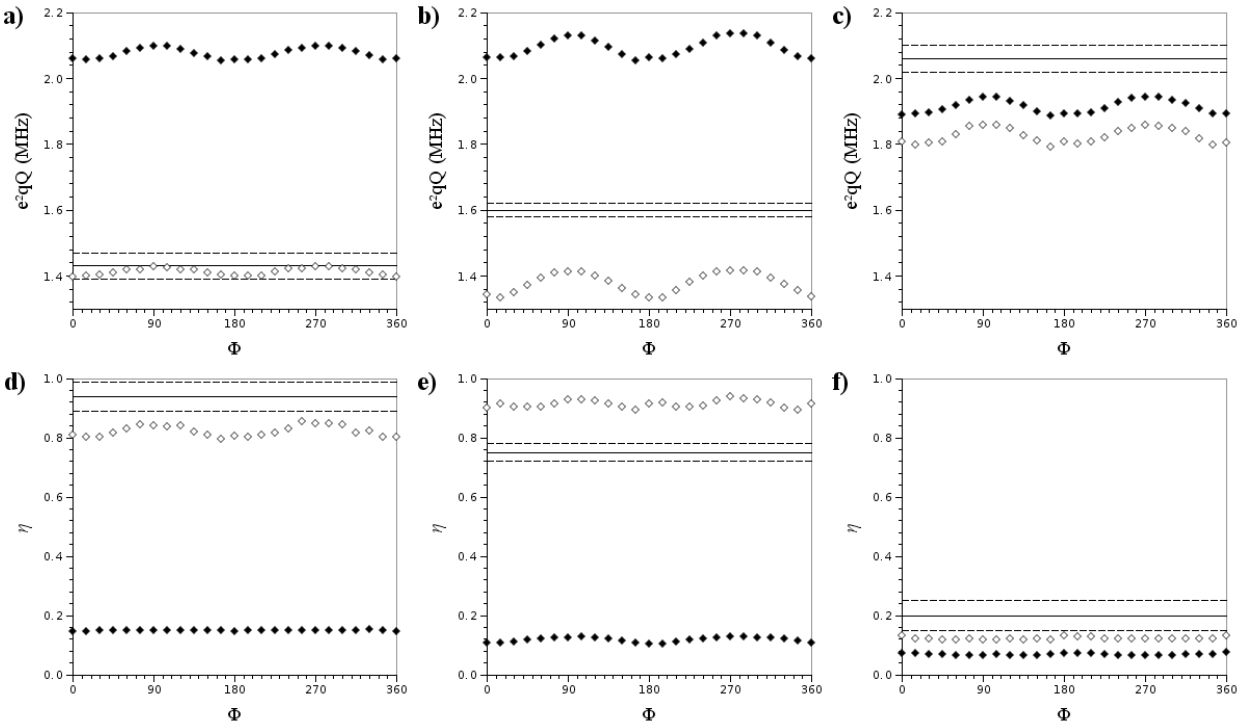

Figure S5. BP calculated NQI, a) – c), and η, d) – f), for the remote nitrogen of the imidazole, 4-methylimidazole and 1-methylimidazole ligands, respectively. The closed, black, symbols are the calculations performed in the absence of a water molecule, while the open symbols are for calculations done with an explicit water molecule in close proximity to the remote nitrogen atom. The solid lines are experimental values and the dashed lines represent the experimentally determined error for each parameter from Ref. 1.

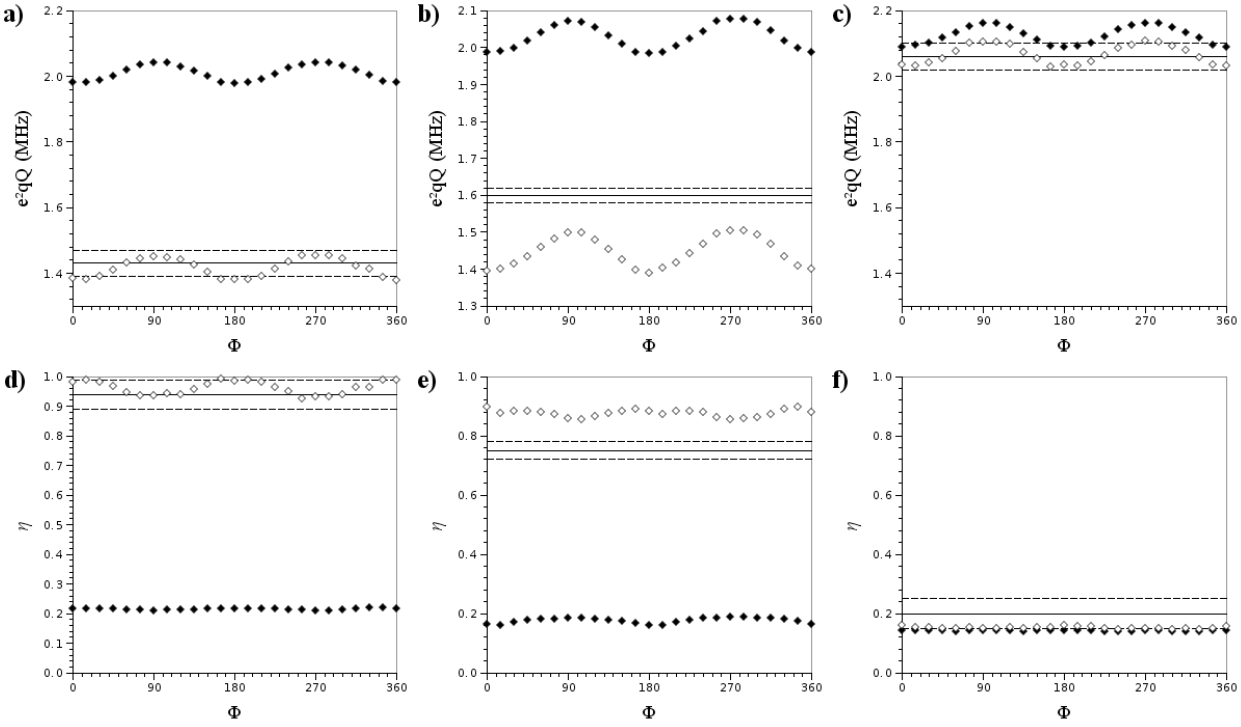

Figure S6. BP calculated NQI, a) – c), and η, d) – f), for the remote nitrogen, utilizing COSMO, of the imidazole, 4-methylimidazole and 1-methylimidazole ligands, respectively. The closed, black, symbols are the calculations performed in the absence of a water molecule, while the open symbols are for calculations done with an explicit water molecule in close proximity to the remote nitrogen atom. The solid lines are experimental values and the dashed lines represent the experimentally determined error for each parameter from Ref. 1.

References:

1. F. Jiang, J. McCracken and J. Peisach, *J. Am. Chem. Soc.*, 1990, **112**, 9035-9044.