# A Comparison of Neural Networks versus Quantum Mechanics for Inorganic Systems

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This paper reports what is, to our knowledge, the first direct comparison of neural networks with quantum mechanical techniques for the prediction of molecular properties for inorganic systems. The molecular properties investigated include prototypical structural (equilibrium bond length, $r_e$), energetic (bond dissociation energy, $D_e$), and vibrational (equilibrium stretching frequency, $\nu_e$) properties used in evaluating new materials. In general, all three molecular properties are predicted more accurately using neural networks as compared to correlated quantum mechanical calculations. The superiority of the neural network approach applies whether the simulation is used to predict a single property or to predict several properties simultaneously.

## INTRODUCTION

Neural networks (NNs) have seen increasing use in chemistry. Gasteiger and Zupan[2] have recently published a monograph on chemical applications of neural networks. NNs show great promise in nonlinear "pattern recognition" applications; some chemical examples include automatic identification of functional groups from molecular spectra and determination of protein structure from amino acid sequence.[2] One application of particular relevance to the present research is the work by Sigman and Rives,[3] who use a neural network to predict atomic ionization potentials using the valence shell electron configuration as input. These applications encouraged us to explore the potential of NNs in a different area of chemistry, i.e., inorganic chemistry, where a main focus of theory and experiment has been the study of patterns in chemical reactivity over the entire periodic table.[4] The underlying chemical patterns inherent in the periodic table arise predominantly from atomic valence electron configurations. It may therefore be possible to use NNs to model the nonlinear patterns which arise in properties of the elements and therefore chemistry when the elements combine to make inorganic molecules. Thus, once sufficiently trained, a NN may be able to provide accurate calculation of properties of inorganic compounds in a very efficient manner.

To the best of our knowledge, this research is the first systematic comparison of neural network versus quantum mechanical methods for prediction of molecular properties for a large series of inorganic species. Diatomics were chosen to test the potential of NNs to model the molecular properties of inorganic systems. Diatomics have several attractive properties for this initial research. First, no structural isomers exist. Future extensions to polyatomics will require information relevant to chemical structure, for example, a connection table; methods for doing this have been developed for organic compounds and should be applicable to inorganics.[2] Second and more importantly, there is a large body of reliable experimental data for chemically diverse species. A cursory glance at the book by Huber and Herzberg[5] reveals almost 200 candidates for which experimental molecular properties are known with sufficient accuracy for the electronic ground states of diatomics.

Molecular properties are traditionally calculated via quantum mechanical (QM) techniques. However, QM methods can be very expensive, even for diatomics, in terms of computer memory, storage, and time requirements.[6] Molecular properties investigated are the equilibrium bond length ($r_e$), stretching frequency ($\nu_e$), and bond dissociation energy ($D_e$) of diatomics (XY), chemical properties that probe the ability of NNs to predict representative structural, vibrational, and enthalpic data, respectively, that a chemist generally employs in evaluating a new material. The quantum calculations described below are not the most accurate to have been performed on many of the diatomics,[6] but they have the advantage that they are all performed at a consistent level of theory to facilitate comparison with NN simulations. The question at the core of this research is the following: Can a simple neural network be trained to outperform, or compete with, traditional quantum chemical approaches for prediction of molecular properties?
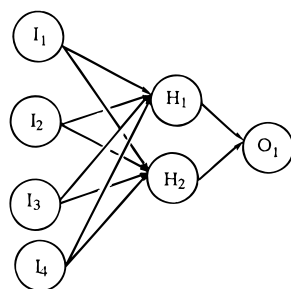
## COMPUTATIONAL METHODS

**1. Neural Network Simulations.** A neural network[2,7,8] is a mathematical construct that models cognitive processes. There are several types of networks; in the present research feedforward neural networks are investigated as these are widely used in chemical applications.[2] A sigmoidal transfer function was used. We focused on three-layer NNs comprised of a single input, hidden and output layer, Scheme 1. A supervised learning algorithm was employed for training. The input data (*vide infra*) are normalized according to eq 1 where $I_n$ = normalized input; $I_x$ = unnormalized input; and $I_{max}$ and $I_{min}$ are the maximum and minimum, respectively.

$$I_n = (I_x - I_{min})/(I_{max} - I_{min}) \qquad (1)$$

Different simulations were initialized with random weights which span a range of values from −0.7 to +0.7. Weights were updated after every epoch. Test calculations were done in which the training patterns were and were not shuffled since some researchers have concluded that the former can forestall "memorization" and improve predictive ability of the NN.[2,7,8] In our research, shuffling did not improve the quality of the results.

The NNs are fully connected, i.e., no short-cut connections between layers were used. Bias units were attached to hidden

**Scheme 1.** A Model 4−2−1 Neural Network



**Table 1.** 13-H-1 Neural Networks Relative to Experiment

| H[a] | $|\Delta_{\nu_e}|$ (cm$^{-1}$) total | $|\Delta_{D_e}|$ (eV) total | $|\Delta_{r_e}|$ (Å) total |
|---|---|---|---|
| 3 | 137 | 0.50 | 0.09 |
| 5 | 75 | 0.40 | 0.06 |
| 8 | 71 | 0.36 | 0.05 |
| 13 | 68 | 0.35 | 0.05 |
| 15 | 66 | 0.31 | 0.05 |
| MCSCF | 100 | 1.11 | 0.14 |

[a] H = number of hidden units. Both testing and training patterns are used in calculating the average absolute error.

and output layers. Because NNs were trained using supervised learning, there was a known output value for each input pattern that can be used to calculate an error function; minimization of the error function was accomplished by the backpropagation (BP) algorithm procedure to adjust the node-to-node weights. The hyperbolic arctangent (atanh) was used as the error function. The BP technique minimizes the error function in an iterative fashion using gradient descent optimization; variations on the backpropagation method can be applied to speed up convergence of the NN simulation. The Quickprop[8] adaptive learning method is employed.[2] For the Quickprop method[8] the maximum step size (called $\mu$ by Fahlman) is 1.75 times the previous step. The Quickprop algorithm is a second-order method which makes the assumption that the error versus weight curve is parabolic for each weight.

In the present research, the number of output nodes was either 1 (prediction of an individual property) or 3 (simultaneous prediction of $r_e$, $\nu_e$, and $D_e$). Following the error in the training and testing sets as a function of epochs (13-13-1 and 13-13-3 NNs were used for this purpose) indicated that the training, testing, and total errors are minimized after $\approx 12\,000 - 13\,000$ epochs, and thereafter little improvement was observed. Similar behavior was observed for both individual and simultaneous predictions of $r_e$, $\nu_e$, and $D_e$. Thus, the results reported below were trained for 15 000 epochs.

A training-to-testing ratio of 3:1 was used.[3,7c] Diatomics were assigned to either the training or testing set randomly. These were then fixed for all subsequent simulations with a given NN topology.

**2. Quantum Mechanical Calculations.** Quantum mechanical calculations were run within the framework of molecular orbital theory using Hartree−Fock and multiconfiguration self-consistent field (MCSCF) wave functions.[9] For closed-shell diatomics the restricted Hartree−Fock (RHF) formalism was employed; for open-shell systems the restricted open-shell Hartree-Fock (ROHF) methodology was used. The GAMESS quantum chemistry program was employed for all quantum mechanical calculations.[10] MCSCF calculations are started using optimized geometries and molecular orbitals obtained from single-configuration RHF/ROHF calculations. Insofar as possible a standard scheme was used for selection of the MCSCF active space to facilitate comparison among a diverse series of compounds. The FORS (full optimized reaction space) approach described by Ruedenberg and co-workers was employed.[9] All possible configurations are considered in the FORS scheme within the limitations of spatial and spin symmetry of the diatomic. In the FORS approach, the active space consists of molecular orbitals derived from atomic orbitals normally considered

as valence and electrons contained therein. For s-block metals and p-block elements the valence orbitals were $n$s and $n$p; for d-block metals the valence orbitals were $n$d and $(n + 1)$s. Finally, for f-block metals the valence orbitals were the 4f orbitals.

All quantum calculations use the effective core potentials developed by Stevens and co-workers.[11] For transition metals the $n$s, $n$p, $n$d, $(n+1)$s and $(n+1)$p electrons were explicitly included, while the remainder are replaced by the ECP. For main group elements the $n$s and $n$p electrons were explicitly included in the quantum calculation. Hydrogen has a −31G basis set. The Stevens ECP scheme has been extensively employed in numerous quantum mechanical studies of chemical compounds incorporating elements from nearly the entire periodic table.[12]

### RESULTS AND DISCUSSION

Thirteen inputs were used to describe each diatomic: atomic number, atomic weight (to study isotope effects), and valence electron configuration (i. e., the number of valence s, p, d, and f electrons) for both atoms, and the overall charge. The NN outputs were the three properties of interest: equilibrium bond length ($r_e$, Å), equilibrium stretching frequency ($\nu_e$, cm$^{-1}$), and bond dissociation energy ($D_e$, eV). NN simulations were run to assess the ability of the NN simulation to predict each property individually and to predict all three properties simultaneously. The data consisted of 150 training patterns and 49 testing patterns.[13] A batch file randomized all initial weights and started each NN simulation. Two thousand separate NN simulations were each run for 15 000 epochs to obtain the best prediction of $\nu_e$, $D_e$, and $r_e$.

The 199 diatomics studied as well as the Quickprop-optimized weights for the different neural network topologies with the best individual or simultaneous predictive ability for equilibrium bond length, stretching frequency, and bond dissociation energy are given in Supporting Information or can be obtained from the authors. The results of the analyses are collected in Table 1. The data are analyzed in terms of the average absolute error, eq 2, between the experimental ($P_{exp}$) and NN-predicted ($P_{NN}$) values, eq 2, of a particular property $P$; $N$ is the total number of samples.

$$\left[\sum_{i=1}^{N} |P_{NN} - P_{exp}|_i\right] \div N = |\Delta_P| \qquad (2)$$

**1. Individual Prediction of $\nu_e$, $D_e$, and $r_e$.** Five different 13-H-1 ($H = 3, 5, 8, 13, 15$) NNs were investigated. Two thousand simulations were run for each topology (each for 15 000 epochs). The results for the simulation which shows the best individual prediction of each of the three properties

**Table 2.** Mean Absolute Differences of Neural Network and Quantum Mechanically Predicted Properties Relative to Experiment[a]

| | $|\Delta_{\nu_e}|$ (cm$^{-1}$) total | $|\Delta_{D_e}|$ (eV) total | $|\Delta_{r_e}|$ (Å) total |
|---|---|---|---|
| 13-13-3 best | 81 | 0.40 | 0.09 |
| 13-13-1 | 68 | 0.35 | 0.05 |
| RHF | 100 | NC[b] | 0.10 |
| MCSCF | 100 | 1.11 | 0.14 |

| | $|\Delta_{\nu_e}|$ (cm$^{-1}$) train | $|\Delta_{D_e}|$ (eV) train | $|\Delta_{r_e}|$ (Å) train |
|---|---|---|---|
| 13-13-3 best | 75 | 0.34 | 0.08 |
| 13-13-1 | 57 | 0.32 | 0.05 |
| RHF | 110 | NC[b] | 0.12 |
| MCSCF | 104 | 1.15 | 0.15 |

| | $|\Delta_{\nu_e}|$ (cm$-1$) test | $|\Delta_{D_e}|$ (eV) test | $|\Delta_{r_e}|$ (Å) test |
|---|---|---|---|
| 13-13-3 best | 99 | 0.57 | 0.11 |
| 13-13-1 | 100 | 0.41 | 0.07 |
| RHF | 70 | NC[b] | 0.06 |
| MCSCF | 88 | 0.99 | 0.11 |

[a] 13-13-3 best is the neural network simulation that yields the best simultaneous prediction of all three experimental quantities (see text). 13-13-1 denotes the neural network simulations with the best _individual_ predictive ability of the molecular properties indicated. Tables with the above simulation results for all diatomics are available from the authors or as Supporting Information. [b] NC = not calculated.

for the combined set of 150 training and 49 testing patterns are given in Table 1. As expected, agreement with experiment is better as the number of hidden units increases. After eight hidden units there is virtually no improvement in predictive ability for the NNs, particularly for $r_e$ and $\nu_e$, although $D_e$ still shows a marginal decrease in $|\Delta_{D_e}|$ up to 13 hidden units. The rest of the discussion in this section will focus on results for 13 hidden units.

**a. 13-13-1 Neural Network Optimized for $\nu_e$.** The 13-13-1 neural network with the best predictive ability with respect to $\nu_e$ for diatomics in the testing set shows an average absolute theory-experiment error ($|\Delta_{\nu_e}|$) of 100 cm$^{-1}$, Table 2; $|\Delta_{\nu_e}|$ for the entire data set (training plus testing) is 68 cm$^{-1}$ for this simulation, yielding $|\Delta_{\nu_e}|$ for the training set of 57 cm$^{-1}$, Table 2. The level of agreement in the NN simulation compares favorably with the results of MCSCF calculations, Table 2: $|\Delta_{\nu_e}| = 100$ cm$^{-1}$ for the entire data set. The MCSCF calculations for the 49 (150) diatomics in the testing (training) set show an absolute average error of 88 cm$^{-1}$ (104 cm$^{-1}$). The NN outperforms the quantum-based MCSCF approach with respect to the total and training patterns.

Single-determinant, RHF calculations yield $|\Delta_{\nu_e}| = 100$ cm$^{-1}$ for the entire set and 70 cm$^{-1}$ (110 cm$^{-1}$) for testing (training) patterns. As RHF values are comparable to those determined using MCSCF wavefunctions, similar conclusions can be made when comparing NN and RHF methods. The RHF $\nu_e$ are too high on average (%$\Delta_{\nu_e}$, eq 3) by 13%, as compared to %$\Delta_{\nu_e} = 4\%$ for the NN simulation. Systematic overestimation of experimental frequencies is characteristic of RHF calculation of vibrational frequencies and is primarily due to neglect of anharmonicity.[14] Thus, one advantage of the NN based methods versus the RHF approach is the lack of any systematic errors with respect to prediction of $\nu_e$ in the former.

$$\left[\sum_{i=1}^{N}(\nu_{NN} - \nu_{exp})_i/(\nu_{exp})_i \times 100\right] \div N = \%\Delta_{\nu_e} \quad (3)$$

**b. 13-13-1 Neural Network Optimized for $D_e$.** The 13-13-1 neural network with the best predictive ability for $D_e$ yields $|\Delta_{D_e}| = 0.41$ eV for the testing set. The training set has $|D_e| = 0.32$ eV, Table 2, for this NN simulation. Thus, $|\Delta_{D_e}| = 0.35$ eV for the entire set of 199 diatomics.

Bond energies are notoriously difficult to calculate accurately using quantum mechanical methods. Since bond dissociation energies are not reliable using single-determinant methods, $D_e$ was not calculated at the Hartree–Fock level of theory. Bond energies of very small species such as diatomics can be calculated to very high precision with more extensive treatments of basis set and electron correlation effects than is employed here, but such quantum calculations are very expensive even for diatomics and of limited utility for larger chemical species.[6] For example, even employing a relatively small active space and basis set, as we have done here, for ZrO ($^1\Sigma^+$) requires over seven hours for determination of the equilibrium bond length. Employing multiconfiguration wavefunctions, and hence including electron correlation in the quantum calculations, yields the following results: $|\Delta_{D_e}| = 1.15, 0.99$, and $1.11$ eV for training, testing, and total patterns, respectively. Thus, comparison with experimental $D_e$, shows the neural network-predicted bond dissociation energies to be more accurate by more than a factor of 2, for both the training and testing sets. It must also be stressed that the NN simulations (2000 different simulations each run for 15 000 epochs) take orders of magnitude less time than determining the bond energies of 199 diatomics using MCSCF methods. Finally, the NN simulation uses simple, easily obtained input and thus does not have the ambiguity which can arise in multiconfiguration calculations from the selection of an active space.

**c. 13-13-1 Neural Network Optimized for $r_e$.** Structural data for chemical compounds are generally easier to calculate than either stretching frequencies and especially bond dissociation energies using quantum methods. The 13-13-1 NN which had the lowest error for $r_e$ yields $|\Delta_{r_e}| = 0.05$ Å for the complete data set, Table 1. For the testing (training) set $|\Delta_{r_e}| = 0.07$ (0.05) Å. MCSCF calculations yield $|\Delta_{r_e}| = 0.11$ Å for the testing patterns and 0.15 Å for the training set, for a combined $|\Delta_{r_e}|$ of 0.14 Å for the entire data set. The 13-13-1 NN optimized for $r_e$ gives better predictive ability, by a factor of 3, when compared to those predicted with MCSCF wavefunctions for the same training set. The NN also outperforms the MCSCF method for the testing set, resulting in overall better predictive ability with respect to $r_e$ for the NN for the entire data set (0.05 Å (NN) versus 0.14 Å (MCSCF)). The 13-13-1 NN simulation also outperforms the RHF calculations ($|\Delta_{r_e}|$ for RHF = 0.10 Å) for all 199 diatomics.

A large part of the error with the QM methods is due to large inaccuracies for systems which are notoriously difficult to describe using Hartree–Fock methods, for example, complexes involving alkali and alkaline earth metals. For example, the MCSCF-predicted bond lengths range from 0.23 Å _too short_ (RbBr) to 0.72 Å _too long_ (Cs$_2$). The former case is consistent with other ECP calculations on the heavier alkali metals[11b,12] and is due to problems arising from inclusion of the outermost core orbitals in the ECP and hence neglect of a significant core-polarization effect. Weakly bound complexes such as alkali metal

**Table 3.** 13-H-3 Neural Networks Relative to Experiment

| $H^a$ | $|\Delta_{\nu_e}|$ (cm$^{-1}$) total | $|\Delta_{D_e}|$ (eV) total | $|\Delta_{r_e}|$ (Å) total |
|---|---|---|---|
| 3 | 229 | 0.99 | 0.20 |
| 5 | 158 | 0.59 | 0.12 |
| 8 | 129 | 0.42 | 0.09 |
| 13 | 81 | 0.40 | 0.09 |
| MCSCF | 100 | 1.11 | 0.14 |

$^a$ H = number of hidden units. Both testing and training patterns are used in calculating the average absolute error.

diatomics generally require more extensive correlation treatments than included in the current MCSCF calculations, e.g., the inclusion of dynamic correlation. Thus, another advantage of neural networks versus quantum methods lies in the fact that prediction of chemical properties by the latter is generally more sensitive to computational difficulties which arise due to the position of the elements in the periodic table, for example, light versus heavy and transition metal versus main group congeners.[12]

**2. Simultaneous Prediction of $\nu_e$, $D_e$, and $r_e$.** A great savings in computational effort would be achieved if one could develop methods to *simultaneously* predict several molecular properties with reasonable accuracy. This goal is not generally achieved with quantum calculations, particularly for larger polyatomics, since separate calculations (sometimes employing different levels of theory), for example, are typically performed to accurately determine metric and energetic data.[12,15] Many researchers employ relatively inexpensive, single-determinant QM calculations to predict geometries; subsequently, higher levels of theory, larger basis sets, or inclusion of electron correlation are used for accurate determination of energetics.[12,15] Neural networks approaches to chemical modeling possess the ability to afford simultaneous prediction of several chemical properties, although this is clearly a greater challenge than prediction of a single property.

**a. Effect of Changing the Number of Hidden Units.** Four different 13-H-3 ($H$ = 3, 5, 8, 13) NNs were investigated. Two thousand simulations were run for each topology (each for 15 000 epochs). The results for the simulation which shows the best simultaneous prediction of all three properties for the complete set of 199 diatomics is given in Table 3. As expected, agreement with experiment is better as the number of hidden units increases. In general, a greater number of hidden nodes ($8 \leq H \leq 13$) were required to yield predictive ability for $\nu_e$ commensurate with MCSCF calculations. For $r_e$ and $D_e$, better predictive behavior was obtained for the NN with relatively few hidden nodes ($3 \leq H \leq 5$). Thus, section 2.b focuses on 13-13-3 results.

**b. 13-13-3 Neural Network Optimized for $\nu_e$, $D_e$, and $r_e$.** The 13-13-3 NN with the best, simultaneous predictive ability for all three diatomic properties ($\nu_e$, $D_e$, and $r_e$) yielded the following average absolute theory-experiment differences: $|\Delta_{\nu_e}|$ = 99 (75) cm$^{-1}$, $|\Delta_{D_e}|$ = 0.57 (0.34) eV, and $|\Delta_{r_e}|$ = 0.11 (0.08) Å for the testing (training) set patterns, Table 2. When the data for the training and testing patterns are combined one obtains the following: $|\Delta_{\nu_e}|$ = 81 cm$^{-1}$, $|\Delta_{D_e}|$ = 0.40 eV, and $|\Delta_{r_e}|$ = 0.09 Å. Since the three properties cover widely different ranges (in the units quoted), the "best" NN simulation was determined by the lowest average error (theory versus experiment for the 49 testing

patterns) for the *normalized* data. To reiterate, MCSCF calculations yielded the following results: $|\Delta_{\nu_e}|$ = 100 cm$^{-1}$, $|\Delta_{D_e}|$ = 1.11 eV, and $|\Delta_{r_e}|$ = 0.14 Å for the entire set of patterns. Hence, for the entire set of 199 diatomics, the 13-13-3 NN outperforms the MCSCF calculations for bond lengths, bond energies, and stretching frequencies.

## SUMMARY AND CONCLUSION

This paper reports what is, to our knowledge, the first direct comparison of neural networks with quantum mechanical techniques for the prediction of molecular properties for inorganic systems. The molecular properties investigated include prototypical structural (equilibrium bond length, $r_e$), energetic (bond dissociation energy, $D_e$), and vibrational (equilibrium stretching frequency, $\nu_e$) properties used in evaluating new materials. In the present research it was found that, in general, NN methods, trained using an adaptive learning technique, outperformed quantum calculations which do (MCSCF) and do not (RHF) include electron correlation. Several interesting results have emerged which are of interest with respect to more general application of neural networks to molecular systems.

(1) There is a considerable reduction in computational effort using NNs since thousands of simulations can be run in the time it takes to run several quantum calculations. The properties evaluated here can often be calculated with great accuracy employing extremely high level quantum calculations. In such a case, the NN approach gains an even bigger advantage in terms of reduced effort and applicability to larger molecules.

(2) The potential of NN methods to *simultaneously* predict several properties is exciting since this is not often achieved with quantum calculations.

(3) Prediction of bond energies is of crucial importance in chemical modeling, for example, catalyst activity. Also, accurate calculation of enthalpic data requires considerable computational expense and is often prohibitive for larger molecules. NNs performed significantly better than QM calculations with respect to calculation of bond energies. Generalization of the current approach to polyatomics could provide an efficient route to estimation of chemical energetics.

(4) A logical extension of the ability to accurately predict bond lengths for diatomics with NNs is prediction of molecular geometries for polyatomics. The study of polyatomic species will require techniques for distinguishing structural isomers. Methods for addressing this issue have been developed for organic compounds and should be applicable to inorganics.[2]

(5) Challenges can arise in application of QM methods to chemical systems incorporating certain families of elements. For example, QM calculations on lanthanide and transition metals typically require more computational effort (larger basis sets and/or more extensive electron correlation) to achieve similar accuracy as compared to lighter main group elements. A potential advantage of NN versus QM methods is that prediction of chemical properties by the latter is generally more sensitive to computational difficulties which arise due to the position of the elements in the periodic table.[12]

Alternative neural network approaches, for example, nonfeedforward networks,[2] employing algorithms such as

Cascade Correlation[16] and pruning[17] to further investigate different NN architectures, recurrent networks,[18] etc. are worthy of investigation, as is comparison with emerging quantum methodologies such as density functional theory[19] which have recently shown much promise in quantum modeling. Furthermore, comparison of neural network approaches with statistical methods is another avenue for further study. The current research indicates that neural networks have considerable interest as an alternative or complement to quantum mechanical techniques for modeling chemical systems, and it is hoped that the present research will encourage further exploration in the application of NNs to inorganic chemistry.

## ACKNOWLEDGMENT

**Supporting Information Available:** Tables of optimized weights, systems studied, and the comparison of experimental molecular properties ($D_e$, $r_e$, $\nu_e$) with those calculated by quantum mechanical and neural network methods are given for all simulations discussed (30 pages). See any current masthead page for ordering and Internet access instructions.

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