Spectral barcoding of quantum dots: Deciphering structural motifs from the excitonic spectra

 ν United Merrian $\frac{1}{2}$ and ν and $\frac{1}{2}$ $E_A = E_A + L_{A} + L_{B} + L_{C}$ Received 1 January 2009; revised manuscript received 200 and 10 200 July 2009- S_α . The conductor \mathfrak{g}_α in high-resolution single-dot spectra and spectra a multitude of \mathfrak{g}_α sharp lines, resembling a barcode, due to various neutral and charged exciton complexes. Here we propose the propose the propose the set of \mathbf{h} \mathcal{S} spectral barcoding method that deciphers structural motifs of dots by using such barcode as input to an artificial-intelligence learning system. Thus, we inverte the common problem problem problem spectra from structure spectra from spectra from spectra from structure spectra from spectra from spectra from spectra from spec by deducing structure from spectra. This approach i- lays the foundation for building a much needed structurespectra understanding for large nanostructures and ii- can guide future design of desired optical features of $Q_{\rm{max}}$ by controlling during growth only those structural motifs that decide given optical features. The decide given optical features optical features optical features. The decide given optical features optical featur

 \ddotsc 10.1103/_{Phys} 0.03532 $\frac{1}{2}$ $\frac{1}{2}$

I. INTRODUCTION

The establishment of relationship between structure and \mathbf{t} spectra in molecules has historically been facilitated by $\mathbf{t} = \mathbb{R} \mathbf{r}$ and \mathbf{t} and \mathbf{t} spectral final final final final final final final field up the specific groups making up the \mathbf{t}_0 molecules. The relationship between the relationship between $\mathbf{1}_{\mathbf{1}}$ and $\mathbf{1}_{\mathbf{2}}$ and $\mathbf{1}_{\mathbf{3}}$ p lines has propelled our understanding of the nature of the natur chemical bond 3,4 3,4 3,4 and for definition for design for design for design for design for design 3,4 of molecules with given properties. We are contained by \mathbf{I}_1 recent \mathbf{I}_2 years[,](#page-5-6) nano- \mathbf{t} such as \mathbf{t} as \mathbf{t}

have emergency of the set of $\mathbf{1}$

serving interesting physical effects resulting from interesting from interesting from interesting from interesting from interesting \mathbf{r}_1 tronic and spin interactions in \mathbb{R}^{12} , \ldots , unlike the case of discrete molecules and crystals, the position of each and every atom in the $\mathbf{t}_i = \mathbf{t}_i \mathbf{t}_i = \mathbf{t}_i \mathbf{t}_i$ and $\mathbf{t}_i = \mathbf{t}_i \mathbf{t}_i$ and known. 13,15 13,15 13,15 Moreover, the association of vibration of 11 fingerprints with specific molecular groups is currently not assume in \mathbf{u} and \mathbf{u} applied to $\mathbf{y} = \mathbf{y} \cdot \mathbf{y}$ at the spectroscopy. Yet, at the heart of most conceived optical application of \mathbf{r}_1 and \mathbf{r}_2 applies light emitting light emitting \mathbf{r}_2 markers, $\frac{16}{10}$ single-photon sources, $\frac{1}{2}$ or solid-state laser[s18](#page-6-3)-17 or solid-state lasers

, \sim 10^4 , 10^6 d() α is not known. Therefore, our proposed is not proposed in approach is somewhat different from the different from the different from the different \mathbf{u} $\mathbf{1}$ chemistry.

II. SPECTRAL BARCODING PROCEDURE

A. Philosophy behind spectral barcoding

 $\mathbf{u} = \mathbf{u} \cdot \mathbf{u}$ emission spectrum, consisting of all lines of all line and the internal behavior international behavior $\mathbf{1}$ be viewed as the $\mathbf{1}$ partial spectral information from the spectra will be denoted by denoted with \mathbf{u}_1 be denoted by here as the $\mathbf{1}_{\{1,2,3\}}$ or $\mathbf{1}_{\{1,3,4\}}$ or $\mathbf{1}_{\{1,3,4\}}$ or $\mathbf{1}_{\{1,3,4\}}$ Spectral marker can be chosen arbitrarily from the emission of \mathbf{r}_1 spectrum but such that it does not depend on uncontrollable \mathbf{l}_1 variables as, e.g., $\mathbf{1}_{\mathbf{z}}$ as, \mathbf{z}_1 and \mathbf{z}_2 are, we chose chosen \mathbf{z}_2 a subsequence of $\mathbf{1}, \mathbf{1}, \mathbf{1}, \mathbf{1}, \mathbf{1}, \mathbf{1}, \mathbf{1}, \mathbf{1}, \ldots$. If $\mathbf{1}$ marker barcode, as shown in Fig. [1](#page-1-0), $\frac{1}{2}$, $\frac{1}{2}$, $\frac{1}{2}$, $\frac{1}{2}$, $\frac{1}{2}$ ways to select a subset as subset as $(1, \ldots, t)$ and $t \in \mathbb{R}$ sequence of lines, e.g., X−2, X−1, XX0 , and X⁰ ; this sequence of emission lines does not depend on the atomic-scale range $\mathbf{1}_{\mathbb{C}}$ randomic-scale random super

for a spectral marker. Perhaps in the future, \mathbf{r}_1 such an approach might open the possibilities of selection \mathbf{r}_i controlling via growth specific structural motifs so as to as t achieve a desired, target spectral behavior of $\mathbf{1}_{\{1,2,\dots,N\}}$ are of similar attenpts in molecular spectroscopy to use \mathbf{t}_i spectroscopy to use the use of use to use spectra and deduce structure. For example, in molecular 1 , 1 , 1 , 1 , 1 chemistry, various forms of spectroscopy have been used to the better \mathbf{t}_i infer structure. In the NMR fingerprints have been structure. In the $N_{\rm eff}$ associated with specific molecular groups using a 1 specific molecular groups using a 1 intelligence methods see e.g., α and β and β and β and β currently available spectroscopy of self-assembled $\mathbf{1}$ spectroscopy of self-assembled $\mathbf{1}$ s ists of electronic spectra only and, furthermore, furthermore, establishing s $\ln \left(1 + \frac{1}{2} \right)$ and special molecular groups and special models are special models.

thal features is currently in \mathbf{u} in actual position of the atoms with such large nanosystem such large nanosystem \mathbf{t}

 $1\,663\,200$ possibilities. We note that achieving the correction of the cor multipliers are contributed in the sequence is not only in the sequence is not only in the sequence is not only in the sequence in the sequence is not only in the sequence in the sequence is not only in the sequence in th $t_{\rm c}$ of the large number of α possible combinations but also but also but also but also but also because it requires the flat rather accurately the electron accurately the electron and hole wave functions. The spectral barcoding method method method method method method method method method does not rely on any specific \mathbf{t}_0 for \mathbf{t}_1 \mathbf{t}_2 \mathbf{t}_3 \mathbf{t}_4 \mathbf{t}_5 \mathbf{t}_7 system, in particular, in particular, except requiring sufficient spectrum sufficient spectrosscopie data $t_{\text{w}} \sim 10$ emission lines-lines is the application of the method to cases where such rich \mathbf{r}_i lengths and and heights $\left[\begin{matrix} 1&1\ 1&1\end{matrix}\right]$, either the growth kind $\left[\begin{matrix} 1&1\ 1&1\end{matrix}\right]$, $\left[\begin{matrix} 1&1\ 1&1\end{matrix}\right]$ netics of \mathcal{A} and \mathcal{A} imposes the limitations or emissions or emissions of \mathcal{A} from the dots $\mathbf{f} = \mathbf{f} + \mathbf{f} + \mathbf{f}$ outside shaded regions in the shaded regions in the shaded regions in the shaded region of \mathbf{f} $\begin{array}{ccc} \textcolor{red}{\mathbf{-1}} \ \textcolor{red}{\mathbf{-2}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-4}} \ \textcolor{red}{\mathbf{-2}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-4}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-4}} \ \textcolor{red}{\mathbf{-5}} \ \textcolor{red}{\mathbf{-4}} \ \textcolor{red}{\mathbf{-5}} \ \textcolor{red}{\mathbf{-1}} \ \textcolor{red}{\mathbf{-1}} \ \textcolor{red$ $\begin{array}{ccc} \textcolor{red}{\mathbf{-1}} \ \textcolor{red}{\mathbf{-2}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-4}} \ \textcolor{red}{\mathbf{-2}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-4}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-4}} \ \textcolor{red}{\mathbf{-5}} \ \textcolor{red}{\mathbf{-4}} \ \textcolor{red}{\mathbf{-5}} \ \textcolor{red}{\mathbf{-1}} \ \textcolor{red}{\mathbf{-1}} \ \textcolor{red$ $\begin{array}{ccc} \textcolor{red}{\mathbf{-1}} \ \textcolor{red}{\mathbf{-2}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-4}} \ \textcolor{red}{\mathbf{-2}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-4}} \ \textcolor{red}{\mathbf{-3}} \ \textcolor{red}{\mathbf{-4}} \ \textcolor{red}{\mathbf{-5}} \ \textcolor{red}{\mathbf{-4}} \ \textcolor{red}{\mathbf{-5}} \ \textcolor{red}{\mathbf{-1}} \ \textcolor{red}{\mathbf{-1}} \ \textcolor{red$

iii) $\prod_{i=1}^k \prod_{i=1}^k \prod_{i=1}^k$ $\mathbf{t}(1,1)$ it to satisfy the sequence of 11 excitonic lines Fig. $\begin{pmatrix} 1 \end{pmatrix}$ $\begin{pmatrix} 1 \end{pmatrix}$ $\begin{pmatrix} 1 \end{pmatrix}$ and $\begin{pmatrix} 1 \end{pmatrix}$ and the requirement for a particular shape now arises see $\left[\begin{matrix} 1 & 1 \end{matrix}\right]$ $\left[\begin{matrix} 1 & 1 \end{matrix}\right]$ $\left[\begin{matrix} 1 & 1 \end{matrix}\right]$, we can define the structure-spectra learning mass $\left[\begin{matrix} 1 & 1 \end{matrix}\right]$, $\left[\begin{matrix} 1 & 1 \end{matrix}\right]$, $\left[\begin{matrix} 1 & 1 \end{matrix}\right]$ chine to decipher the structural content of the spectral bar-spectral bar-spectral barcode determining the \mathbf{t}_1 is next applied to the BDRA is next applied to the \mathbf{t}_2 remaining excitonic sequence, searching for secondary structure for secondary structure to position that determines the position of \mathbf{t}_1 relative to \mathbf{t}_2 $\frac{1}{2}$, $\frac{1}{2}$ redshifted relative to $\frac{1}{2}$ redshifted relative to $\frac{1}{2}$

and μ be the μ and μ an

 ${\bf t}$ and artificial matrix spectral matrix ${\bf t}$ artificialintelligence learning system bayesian data reduction bayesian data reduction algo- \mathbf{r} in \mathbf{r} and \mathbf{r} in \mathbf{r} and $\$ structural motif i- we establish the missing structural basis for ζ spectroscopy, creating a beginning a beginning of a beginning of a different of ζ spectral features vs $\mathbf{1}$ in $\mathbf{1}$ lines for the design of design of $\mathbf{1}$ for $\mathbf{1}$ \math controlling during growth simple structural motifs, rather \mathbf{r}_i $t_{\rm{max}}$ at $t_{\rm{max}}$ at the control the control the control theorem is the complete structure of the complete structure of the control theorem is the control theorem in the control theorem is the control theorem in the large nanosystems.

ACKNOWLEDGMENTS

The authors thank \mathbf{P}_{max} and \mathbf{P}_{max} and \mathbf{P}_{max} and \mathbf{P}_{max} $t_{\rm{max}}=1,1,1,\ldots, n$ and $t_{\rm{max}}=1,1,\ldots,1,1,1$ Fig. [1](#page-1-0) - and R. J. Warburton for discussions and comments on the paper. This work was funded by the U.S. Department was funded by the U.S. Department of U of Energy, Office of Science under NREL Contract $\mathbf{1}_{\mathcal{A}}$, $\mathbf{1}_{\mathcal{A}}$, $\mathbf{1}_{\mathcal{A}}$ $36.0 \t230$

APPENDIX A: ROLE OF ATOMIC-SCALE RANDOMNESS

 \textbf{I}_C , as really allows are usually allowed by structure \textbf{I}_C tural characterization techniques. $\mathbf{14}$ The fact $\mathbf{14}$ $\mathbf{14}$ $\mathbf{14}$ $\mathbf{14}$ $\mathbf{14}$ $\mathrm{I}_{\tau-1,\tau}$, in $\mathrm{I}_{\tau-1}$ is a set of possible spatial configurations, 35 σ , and each can have distinct property of 1 σ).^{[36](#page-6-7)} **t t**₁, **i**₁ **d**₁ **t**₁₁ **e e**ch **i**₁ **c e e i** from an ensemble will have interesting with $\mathbf{1}$ controlled . We have a finite value of 1 and α and 1 and α and 36 however different random realizations $\lim_{n\to\infty} \log\log n$. In fluence emission lines in $\lim_{n\to\infty} \log\log n$ of several multiplexcitonic transitionic transitions. Here we interference $\mathbf{H}_\ell(\mathbf{t},\mathbf{t},\mathbf{t})$ different RRs influence distances between $\mathbf{1}_{\mathcal{A}}$ and $\mathbf{1}_{\mathcal{A}}$ and $\mathbf{1}_{\mathcal{A}}$ but not the sequence of lines on the example of 1 and \times $\frac{2}{\pi}$ and x^{1} transitions. Our results are shown in Fig. [4](#page-5-10) μ and $\mathbf{1}$ in determining 1 and 25 nm, and 2 nm, and I_1 –60%].

APPENDIX B: MANY BODY PSEUDOPOTENTIAL CALCULATIONS

 $\mathbf{F}_\mathrm{m} = \mathbf{t} + \mathbf{y} + \mathbf{y} + \mathbf{y}$, and composition of a \mathbf{y} we are a \mathbf{y} for $\mathbf{r}=\mathbf{r}=\mathbf{r}=\mathbf{r}=\mathbf{r}=\mathbf{r}=\mathbf{r}=\mathbf{r}=\mathbf{r}$

91, 263105, 200).

- 14 J. Stangl, V. Holy, and G. Bauer, Rev. Mod. 25 2004).
- 15M. Bruls, J. W. A. M. Vugs, P. M. Koenraad, H. W. M. Salemnik, J. Wolfen, M. Wolter, M. Wolfen, F. Long, and S. P. Skolnick, F. Long, and S. P. Long, and S. P. Long, and S.
- \ldots **1**, **1**, **1**, **1**, **1**, **81**, 1 0 **4** 2002). $\frac{16}{16}$, \ldots , $\frac{1}{16}$, $\frac{1}{16}$, \ldots $\frac{1}{16}$, $\frac{1}{16}$, \ldots $\frac{1}{16}$, \ldots $\frac{1}{16}$, \ldots
- . ._. . **11** 1₁ 1 . 1 . 19, 11 1, 2006).
- 1 and $\ket{\mathfrak{l}}$. Pelton, $\ket{\mathfrak{r}}$ in $\ket{\mathfrak{r}}$ and $\ket{\mathfrak{r}}$ in $\ket{\mathfrak{r}}$ in $\ket{\mathfrak{r}}$. So that
- **Plant, and Y. Yamamoto, Phys. Rev. 2002. th. 89**, 233602, 2002).
- 1 , and that is, i.e. and the state of the state of the 1 Grundmann $\mathbf{1}$ and $\mathbf{1}$ $\mathbf{U}_\mathbf{A}$, $\mathbf{1} \cdot \mathbf{1} \dots$ **tt. 30**, 1416 1 4).
- $\frac{1}{2}$, methods for structural characterization are structural characterization are still characterization are still not accurate enough to reveal the full atomic-scale structure. For \mathbf{r}_i example, one of the leading characterization methods, \mathbf{t}_1 and \mathbf{t}_2 requires as input a good guess for the shape and composition \mathbf{r}_1 profile of the dot where the dotation is structural determination in the ensuing structural determination is determined as a set of the ensuing structural determination is determined as a set of the ensuing structural det then performed and validated only with a neighborhood of $\mathbf{1}\oplus\mathbf{1}$ and $\mathbf{1}\oplus\mathbf{1}$ and $\mathbf{1}\oplus\mathbf{1}$ $s_1, s_2, \ldots, s_{n-1}$ situation is the situation is strongly diffraction in the situation is with x_1
- \mathcal{R} (and intensity distributions in the reciprocal space space \mathbf{r} in the reciprocal space are recorded. From these records in the recorded maps using initial guesses of \mathbf{r}_i \mathcal{O}_∞ shape and composition profile, and performing strain calculated s
- $\lfloor \textbf{t}_1 \rfloor, \textbf{t}_{\text{out}} \rfloor = \lfloor \textbf{t}_1 \rfloor, \ldots, \lfloor \textbf{t}_n \rfloor = \lfloor \textbf{t}_1 \rfloor, \ldots \lfloor \textbf{t}_n \rfloor,$ transmission electron microscopy-based techniques are currently assumed to \mathbf{u}_i unable to go beyond qualitative description of QD compositional l_{\blacktriangledown} ([13](#page-5-9)).
- $\frac{20}{20}$ a. A. $\frac{1}{2}$ become and R. A. Burton, private communication, private communication, private communication \therefore **t** 200).
- $^{21}(\ ,\ ,\quad ,\ ,\ ,\ \ldots)$, $_{1}$. We arder \mathbb{L} , $_{\sim}$. In action is \mathbb{L} , $\zeta_{\rm C}$, and $\zeta_{\rm C}$ are positive and $\Gamma_{\rm C}$, and $\zeta_{\rm C}$ and $\zeta_{\rm C}$ \therefore **76**, 235304 200.
- 22 , bayer, $\mathbf{1}$, $\mathbf{1}$ $\mathbf{t}_{\mathbf{l}}$ **405**, 23 2000).
- 2^3 . **A.** $\overrightarrow{11}$, $\overrightarrow{1}$, $\overrightarrow{2}$, $\overrightarrow{1}$, $\overrightarrow{1}$

 12130 , $\frac{1}{2}$ 200).

- 2^4 . \ldots , \ldots , 1 . \ldots , 11 . **23**, 35 , 1
- 25 . $$, $$ **1**, $$, 2005).
- $26(N-1)$, we will note that spectral spectral-barcode features sp the spectra that are altered by using different area of the spectra that are altered by using different by using different are altered by using different by using different area of the different by using different are dif \mathbf{t} -state randomness. For example, a dot made of \mathbf{t}
- $+$ Inas can manifest different configurations of \mathcal{A}_1 on cations of \mathcal{A}_2 sites for the fixed geometry and composition but calculation but calculations of \mathbf{u}_i $\begin{array}{ccc} 11 & 11 & 101 \end{array}$