Evolution in time of an *N*-atom system. II. Calculation of the eigenstates

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(Received 17 July 2003; published 23 January 2004)

We calculate the energy eigenvalues and eigenstates corresponding to coherent single and multiple excitations of a number of different arrays of *N* identical two-level atoms (TLA's) or qubits, including polygons, ''diamond'' structures, polygon multilayers, icosahedra, and dodecahedra. We assume only that the coupling occurs via an exchange interaction which depends on the separation between the atoms. We include the interactions between *all* pairs of atoms, and our results are valid for arbitrary separations relative to the radiation wavelength.

DOI: 10.1103/PhysRevA.69.013815 PACS number(s): 42.50.Fx, 36.40.Mr

In this paper, we calculate the eigenstates of some regular geometrical arrangements of *N* identical (coherently excited) two-level quantum systems. Such systems are known as qubits to the quantum information community and as two-level atoms (TLA's) to the quantum optics/spectroscopy community. The coherent excitation of identical TLA's has long been of interest to spectroscopists, in connection with the theory of molecular excitons [1] for example, or the phenomenon of superradiance [2]. More recently, the interest has been in connection with the optical properties of molecular clusters or aggregates [3], many of which properties are believed to be related to the coherent interaction of the aggregates with the radiation field. At the same time, multiparticle entangled states of qubits have become an active area of study in the field of quantum information theory. The results presented here are of relevance to this community in the studies of decoherence-free subspaces [4] and investigations into the entanglement properties of rings of qubits [5].

We emphasize the complete generality of the majority of the results obtained herein: our results are applicable to all systems in which excitation is exchanged between the pairs of interacting TLA's. Such exchange interactions occur widely: For example, our theory is applicable to systems in which the coupling is via a spin-exchange interaction, or via a retarded dipole-dipole (quadrupole-quadrupole) interaction, such as exists in coherent dipole [6] (quadrupole [7]) radiative excitation of atoms or molecules. Our calculations are valid for arbitrary distances relative to the radiation wavelength, and we do *not* make the common approximation of including only nearest-neighbor interactions, but rather we diagonalize the full Hamiltonian: This is important because for many physically realistic systems the coupling between nonnearest neighbors can exceed that between adjacent atoms.

The eigenstates of regular polygons are calculated in Sec. I. In Sec. I A, we begin by reviewing the calculation of the eigenstates for single ($n=1$) excitations of a system of *N* TLA's arranged at the vertices of a regular polygon and interacting via an exchange interaction, valid for arbitrary *N*. In Sec. I B we present a method for calculating the eigenstates for double ($n=2$) excitations of the system, also for arbitrary *N*. Finally in Sec. I C, we outline the calculation of the triplet ($n=3$) eigenstates for $N=6$ and 7, and present in Tables I–III the complete set of eigenstates for all regular polygons up to and including $N=6$; results for $N=7$ are available upon request.

In Sec. II, we adapt the methods of Sec. I to study other structures: ''diamonds'' and ''pyramids'' in Sec. II A, polygon multilayers in Sec. II B, icosahedra in Secs. II C and II D, and dodecahedra in Sec. II E. As discussed in [8], the optical activity or inactivity of the eigenstates in absorption, as well as their total decay rates in emission, is immediately evident in the imaginary parts of their eigenvalues. For quantum information theorists, there is special interest in these total decay rates in order to identify particularly long lived states, which might be useful for encoding quantum information. To quantum information theorists, these are known as ''decoherence-free'' states, and to spectroscopists as ''subradiant'' states [9]. In general, complete subradiance exists only in the small sample limit, when distance effects are ignored. Since our calculations contain the complete distance dependence, they can be used to examine deviations from the ''long wavelength'' or ''equal collective decoherence'' assumption commonly made in the theory of decoherence-free subspaces.

In the spectroscopy community, the study of collective atomic phenomena is many years old, beginning with Dicke's pioneering article [6]; for the early work, see [2,11], and references therein. A detailed study of the cooperative emission by a fully-excited system of 3 identical atoms in some specific geometrical configurations was performed by

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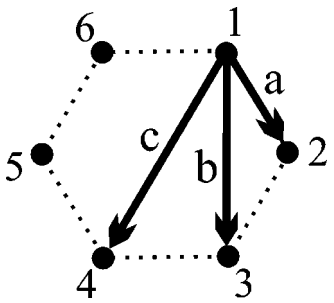

FIG. 1. A regular hexagon of interacting TLA's.

Richter [10], while the complete eigenstates for two- and three-atom systems of arbitrary geometrical arrangement can be found in Ref. [11]. The single-excitation eigenstates of linear chains were presented in [12], of two-dimensional arrays in [13], and of rings and regular polygons in [14]. Single and double excitations of regular polygons in the long-wavelength limit were considered by Spano and Mukamel [3]; however, they included in their calculations only nearest-neighbor interactions, so that our energy eigenvalues and eigenstates differ considerably from theirs.

I. REGULAR POLYGONS

We consider systems of N identical TLA's located at positions $\mathbf{R}_i$, each with ground state $|a_i\rangle$, excited state $|b_i\rangle$ and transition frequency ω_0. The atomic Hamiltonian is then given by

$$H_A=\sum_{i=1}^{N}\hbar\omega_0|b_i\rangle\langle b_i|.$$

Henceforth, we will label states in the ''computational'' basis, i.e., the bare noninteracting states, according to which atoms are excited therein; for example, the state of the $N=5$ system in which atoms 2 and 5 are excited is written as $|B_{25}\rangle\equiv|a_1\rangle|b_2\rangle|a_3\rangle|a_4\rangle|b_5\rangle$. The state with all atoms in the state $|a\rangle$ ($|b\rangle$) is denoted by $|G\rangle$ ($|E\rangle$).

The generic (excitation-) exchange interaction Hamiltonian of the TLA's is given by

$$H_{int}=\sum_{\substack{i,j=1\\ i\neq j}}^{N}\hbar\Omega_{ij}S_i^+S_j^-,$$

where S_i^+ and S_i^- are the raising and lowering operators of atom i. The sole assumption we make regarding the interaction potential Ω_{ij} is that it is a function only of the separation between atoms i and j, $\mathbf{R}_{ij}=\mathbf{R}_i-\mathbf{R}_j$. We focus in this section on atoms arranged at the vertices of regular polygons, and number them sequentially around the polygon (see Fig. 1). For nearest-neighbor atoms, we define $\Omega_{i,i\pm1}=a$; similarly, $\Omega_{i,i\pm2}=b$; for N atoms there are $\lfloor N/2\rfloor$ characteristic interactions, which we label sequentially alphabetically. For use with the master equation, we will include in the full Hamiltonian the free-atom radiation damping $H_d=\Sigma_{i=1}^{N}\hbar\gamma|b_i\rangle\langle b_i|$ as well, and allow the Ω_{ij} to be complex—their exact expressions are given in [8]. As discussed there, the real part of Ω_{ij} is the interatomic interaction energy, and the imaginary part the interatomic damping. [In many systems as well an interaction exists between nearest neighbors (e.g., due to atomic overlap) *in addition* to the electromagnetic exchange interaction which occurs between all pairs. These forces too can be included trivially in our analysis, simply by incorporating them into the nearest-neighbor interaction $\Omega_{i,i\pm1}=a$.] This results in a non-Hermitian Hamiltonian, with complex eigenvalues. As demonstrated in [8], the real part of the eigenvalue gives the shift of energy of the state due to local field effects, and the imaginary part gives its total decay constant (or inverse lifetime).

The full Hamiltonian to be diagonalized is represented by a $2^N\times2^N$ matrix. Fortunately, it is block-diagonal in structure, breaking up into a series of submatrices, in each of which the coupled subsets of states all have the same number n of excited TLA's. The submatrices are of dimension $\binom{N}{n}$, and the submatrix for n excited atoms is the same as that for $N-n$ excited, a general property of exchange interactions; this halves the amount of work we must do (and consequently we tabulate results only for $n=1,...,\lfloor N/2\rfloor$). The $n=0$ and $n=N$ eigenstates are just $|G\rangle$ and $|E\rangle$, respectively.

A. $n=1$: Single excitation eigenstates

The single excitation (or $n=1$) eigenstates of a system of N TLA's arranged at the vertices of a regular polygon were calculated years ago [14], guided by the symmetry of the system under rotation about an axis perpendicular to the polygon plane; the $n=1$ eigenvalues and eigenstates for $N=1-6$ are tabulated there. Here we rewrite these calculations in a notation which allows us to extend them to states containing higher numbers of excited TLA's, using the case of $N=5$ as an example.

In the subspace spanned by the basis vectors $\{|B_1\rangle,|B_2\rangle,...,|B_5\rangle\}$, the matrix to be diagonalized has the form

$$M^{(1)}=\begin{pmatrix}0&a&b&b&a\\a&0&a&b&b\\b&a&0&a&b\\b&b&a&0&a\\a&b&b&a&0\end{pmatrix}.$$

We introduce the matrix P, a generator of the 5-dimensional representation of C_5 (the cyclic group of order 5):

$$P=\begin{pmatrix}0&1&0&0&0\\0&0&1&0&0\\0&0&0&1&0\\0&0&0&0&1\\1&0&0&0&0\end{pmatrix}. \tag{1}$$

The eigenvalue equation of P is given by

$$Pu_{(v)}=\lambda^v u_{(v)},$$

TABLE I. Eigenvalues and eigenvectors of 5 TLA's arranged on the vertices of a regular pentagon.

n	v	Eigenvalues	Eigenvectors
1	1,4	$2(c_2a-c_1b)$	$(c_2,c_4,c_6,c_8,c_{10});\ (s_2,s_4,s_6,s_8,s_{10})$
	2,3	$2(-c_1a+c_2b)$	$(c_4,c_8,c_{12},c_{16},c_{20});\ (s_4,s_8,s_{12},s_{16},s_{20})$
	5	$2a+2b$	(1,1,1,1,1)
2	1,4	$E_\pm(-c_1a,c_2b)$	$(c_2,-c_1,-c_1,c_2,1;\ v_\pm c_2,v_\pm,v_\pm c_2,-v_\pm c_1,-v_\pm c_1)$
			$(-1,-2c_2,2c_2,1,0;\ v_\pm,0,-v_\pm,-2v_\pm c_2,2v_\pm c_2)$
	2,3	$E_\pm(c_2a,-c_1b)$	$(-c_1,c_2,c_2,-c_1,1;\ -w_\pm c_1,w_\pm,-w_\pm c_1,w_\pm c_2,w_\pm c_2)$
			$(-2c_2,1,-1,2c_2,0;\ 2w_\pm c_2,0,-2w_\pm c_2,w_\pm,-w_\pm)$
	5	$E_+(a,b)$	$(1,1,1,1,1;\ u,u,u,u,u)$
		$E_-(a,b)$	$(u,u,u,u,u;\ -1,-1,-1,-1,-1)$
		$c_j=\cos(j\pi/5)$	$u=G_+(a,b)$
		$s_j=\sin(j\pi/5)$	$v_\pm=G_\pm(-c_1a,c_2b)$
		$F(\alpha,\beta)=\sqrt{5(\alpha+\beta)^2-4\alpha\beta}$	$w_\pm=G_\pm(c_2a,-c_1b)$
		$E_\pm(\alpha,\beta)=\alpha+\beta\pm F(\alpha,\beta)$	$G_\pm(\alpha,\beta)=[\alpha-\beta\pm F(\alpha,\beta)]/2(\alpha,\beta)$

where $\lambda\equiv e^{2\pi i/5}$, $u_{(v)}=(\lambda^v,\lambda^{2v},\lambda^{3v},\lambda^{4v},\lambda^{5v})$, and $v=1,...,5$. We define the polynomial $M(x)=a(x+x^4)+b(x^2+x^3)$, in terms of which $M^{(1)}=M(P)$. Since $M^{(1)}$ is a sum of powers of P, the eigenvectors $u_{(v)}$ of P will be eigenvectors of $M^{(1)}$ as well, and we write the eigenvalue equation

$$M^{(1)}u_{(v)}=m_{(v)}u_{(v)},$$

where the eigenvalues $m_{(v)}=M(\lambda^v)\equiv G^{(1)}_{(v)}+iF^{(1)}_{(v)}$. There is 1 nondegenerate eigenvalue corresponding to $v=5$, $m_{(5)}=2a+2b$, and $(5-1)/2$ degenerate pairs of eigenvalues, corresponding to roots which are complex conjugates of each other: $\lambda^v=(\lambda^{5-v})^*$. The eigenvector corresponding to $m_{(5)}$ is simply $u_{(5)}=(1,1,1,1,1)$. For the eigenvectors corresponding to the degenerate pairs of eigenvalues, we choose the real linear combinations of $u_{(v)}$ and $u_{(5-v)}$,

$$U^{(+)}_{(v)}=\frac{1}{2}(u_{(v)}+u_{(5-v)}),$$

$$U^{(-)}_{(v)}=\frac{1}{2i}(u_{(v)}-u_{(5-v)}).$$

Together with $u_{(5)}$, these form an orthogonal basis set for the $n=1$ subspace. They are listed in Table I.

B. $n=2$: Double excitation eigenstates

1. Odd values of N

We continue with the example of $N=5$ to demonstrate how to calculate the $n=2$ (biexciton) eigenstates for general odd values of N. The subspace corresponding to $N=5$, $n=2$ has 10 basis states, which we take in the order $\{|B_{12}\rangle,|B_{23}\rangle,...,|B_{51}\rangle;|B_{13}\rangle,|B_{24}\rangle,...,|B_{52}\rangle\}$.

If we define the four polynomials $M_{11}(x)=b(x+x^4)$, $M_{12}(x)=a(x^4+x^5)+b(x+x^3)$, $M_{21}(x)=a(x+x^5)+b(x^2+x^4)$ and $M_{22}(x)=a(x^2+x^3)$, then the interaction can be represented by the 10×10 matrix,

$$M^{(2)}(P)\equiv\begin{pmatrix} M_{11}(P) & M_{12}(P) \\ M_{21}(P) & M_{22}(P)\end{pmatrix}.$$

Thus, $M^{(2)}$ is partitioned into a 2×2 array of square submatrices, each of dimension 5×5. The ability to write the matrix in this form is directly due to the ordering of the basis vectors, which allows the rotational symmetry of the pentagon to be reflected in each of the submatrices. It is easy to show that for any odd value of N, $M^{(2)}$ can be partitioned in this way into an array of $(N-1)/2\times(N-1)/2$ square submatrices, each of dimension $N\times N$. This results in a dramatic simplification of the problem, for instance here we need diagonalize only a 2-dimensional matrix instead of the original 10-dimensional one.

As with the $n=1$ case discussed above, each matrix $M_{ij}(P)$ is a linear combination of P and its powers, and therefore has the eigenvalue equation

$$M_{ij}(P)u_{(v)}=M_{ij}(\lambda^v)u_{(v)},$$

where λ^v and $u_{(v)}$ are the eigenvalues and eigenvectors of P. In order to obtain the eigenvalues and eigenvectors of $M^{(2)}$, we first solve the eigenvalue equation

$$M(x)V(x)=\mu(x)V(x),$$

where $V(x)$ is an eigenvector and $\mu(x)$ an eigenvalue of the two-dimensional matrix $M(x)$. The solutions are easily found to be

$$\mu^\pm(x)=\frac{1}{2}[M_{11}(x)+M_{22}(x)\pm R(x)],$$

where

$$R(x)=\sqrt{(M_{11}(x)-M_{22}(x))^2+4M_{12}(x)M_{21}(x)}$$

and

TABLE II. Eigenvalues and eigenvectors of a single pair ($N=2$), equilateral triangle ($N=3$), and square array ($N=4$) of TLA's.

N	n	v	Eigenvalues	Eigenvectors
2	1	1	$-a$	$(-1,1)$
		2	a	$(1,1)$
3	1	1,2	$-a$	$(-1,-1,2)$; $(1,-1,0)$
		3	$2a$	$(1,1,1)$
4	1	1,3	$-b$	$(0,-1,0,1)$; $(1,0,-1,0)$
		2	$-2a+b$	$(-1,1,-1,1)$
		4	$2a+b$	$(1,1,1,1)$
	2	1,3	0	$(0,-1,0,1;0,0)$; $(1,0,-1,0;0,0)$; $(0,0,0,0;1,-1)$
		2	$-2b$	$(-1,1,-1,1;0,0)$
		4	$b\pm R$	$(1,1,1,1;x_{\pm},x_{\pm})$
			$R=\sqrt{b^2+8a^2}$	$x_{\pm}=(-b\pm R)/2a$

$$V^{\pm}(x)=\begin{pmatrix} M_{11}(x)-M_{22}(x)\pm R(x) \\ 2M_{21}(x) \end{pmatrix}.$$

The eigenvalues and eigenvectors of $M^{(2)}$ can then be shown by direct substitution to be $\{\mu^{\pm}(\lambda^{v})\}$ and

$$U^{\pm}_{(v)}=V^{\pm}(\lambda^{v})\otimes u_{(v)}=\begin{pmatrix} V^{\pm}_{1}(\lambda^{v})u_{(v)} \\ V^{\pm}_{2}(\lambda^{v})u_{(v)} \end{pmatrix},$$

where $v=1,...,5$. As with $n=1$, for degenerate eigenvalues we form the real linear combinations of the eigenvectors; the complete orthogonal basis set is listed in Table I.

In general, the eigenvalue equation for the $n=2$ excitations of any odd-N array of TLA's is solved in the same way.

(i) The interaction matrix $M^{(2)}$ is partitioned into an array of square submatrices, each of dimension $N\times N$.

(ii) The eigenvalue equation of matrix $M_{ij}(P)$ is solved, where P is the $N\times N$ matrix analogous to Eq. (1).

(iii) The eigenvalue equation is solved for the corresponding $(N-1)/2\times(N-1)/2$ matrix $M(x)$, yielding eigenvalues $\{\mu_{(i)}(x)\}$ and eigenvectors $\{V_{(i)}(x)\}$.

(iv) The eigenvalues and eigenvectors of $M^{(2)}$ are then given by $\{\mu_{(i)}(\lambda^{v})\equiv G^{(2)}_{(vi)}+iF^{(2)}_{(vi)}\}$ and $\{U_{(vi)}=V_{(i)}(\lambda^{v})\otimes u_{(v)}\}$, where $v=1,...,N$, $i=1,...,\lfloor(N-1)/2\rfloor$, $\lambda=e^{2\pi i/N}$, and the vectors $\{u_{(v)}\}$ are the eigenvectors of the matrix P corresponding to the N-sided polygon.

2. Even values of N

The calculations for the $n=2$ energies and eigenstates for even values of N cannot be described (or performed) so succinctly. This is due to the fact that $(N-1)/2$ is an odd half-integer. As a result, the matrix for $M^{(2)}$ consists of 2 parts: an inner "core" of $\lfloor(N-1)/2\rfloor\times\lfloor(N-1)/2\rfloor$ square submatrices, each of dimension $N\times N$, plus an outer section of $N/2$ extra columns to the right and rows at the bottom of the core. For example, the $n=2$ interaction matrix for $N=4$ is given by

$$M^{(2)}=\begin{pmatrix} 0 & b & 0 & b & a & a \\ b & 0 & b & 0 & a & a \\ 0 & b & 0 & b & a & a \\ b & 0 & b & 0 & a & a \\ a & a & a & a & 0 & 0 \\ a & a & a & a & 0 & 0 \end{pmatrix},$$

with an inner core matrix $M(P)=b(P+P^3)$, where P is now the 4-dimensional analogue of Eq. (1).

The calculations are performed in the following manner; we illustrate the general procedure with the example of $N=4$.

(i) The energy eigenvalues and vectors of the "core" matrix are obtained, in exactly the same way as described in the previous section for odd values of N.

For the case of $N=4$, the eigenvectors of $M(P)$ are the same as those of $N=4$, $n=1$, which in turn are the same as those of P. They appear in Table II.

(ii) These eigenvectors are then divided into 2 groups, according to their symmetry or antisymmetry. The vectors $(1,1,...,1,1)$ and $(1,-1,1,-1,...,1,-1)$ are always eigenvectors, the former symmetric, the latter antisymmetric; the remainder are classified according to their symmetry under rotations of π about the symmetry axis.

In the case of $N=4$, three of these eigenvectors (those corresponding to $v=1$, 2, and 3 as listed in Table II) are antisymmetric, while that corresponding to $v=4$ is symmetric.

(iii) The antisymmetric eigenvectors are appended with $N/2$ 0's; the resulting vectors are eigenvectors of $M^{(2)}$, and the corresponding energies are found by direct substitution.

In the case of $N=4$, by appending two 0's to the ends of the antisymmetric vectors, we obtain the following three eigenvectors of $M^{(2)}$:

$$U_{(1)}=\begin{pmatrix}1\\0\\-1\\0\\0\\0\end{pmatrix},\quad U_{(2)}=\begin{pmatrix}-1\\1\\-1\\1\\0\\0\end{pmatrix},\quad U_{(3)}=\begin{pmatrix}0\\1\\0\\-1\\0\\0\end{pmatrix}.$$

The corresponding eigenvalues are found by substitution. By symmetry, we see that a fourth (antisymmetric) eigenvector of $M^{(2)}$ is $U=(0,0,0,0,1,-1)$.

(iv) The symmetric eigenvectors are extended into the rest of the $n=2$ subspace in a symmetric fashion.

For the example of $N=4$, the remaining 2 eigenvectors are found from the symmetric eigenvector $u_{(4)}=(1,1,1,1)$ of $M^{(1)}$. We substitute into the eigenvalue equation for $M^{(2)}$ the trial vector $U=(1,1,1,1,x,x)$, obtaining 2 (independent) equations for x and the eigenvalues μ:

$$2b+2ax=\mu,\quad 4a=\mu x.$$

These have the solutions

$$\mu^{\pm}=b\pm R,\quad x^{\pm}=\frac{4a}{b\pm R},$$

where $R=\sqrt{b^2+8a^2}$. This completes our set of 6 eigenvectors of $M^{(2)}$. They are listed together with their corresponding eigenvalues in Table II.

A number of simple properties are immediately evident in these eigenvalues and vectors. For example, in the long-wavelength limit only one $n=1$ state is optically active in absorption and emission, and it is superradiant, having an eigenvalue whose imaginary part$\rightarrow N\gamma$; the $N-1$ other single-excitation states are subradiant, with the imaginary parts of their eigenvalues$\rightarrow 0$. In general, there are $\lfloor N/2\rfloor$ (optically active) $n=2$ states which can be excited sequentially via two-photon absorption from the ground state. In general too, the $n=2$ states decay into $n=1$ states (although these in turn may be subradiant); however, for even-N polygons, there is (at least) one $n=2$ state which is itself completely subradiant. We point out that the $N=4$, $n=2$ eigenstates are the first which depend on the actual strength of the interaction, and not merely on its symmetry. In Fig. 2, we illustrate the distance dependence of their decay rates. In the long-wavelength limit, three of the eigenstates have their decay rates unchanged from the noninteracting value of 2γ, and one state is superradiant, with an asymptotic value of 5.930γ ([8], Table III). The remaining two states are subradiant: One shows weak optical activity, with asymptotic decay rate 0.070γ, and one is *completely* subradiant, with decay rate $\rightarrow 0$; thus, this state is of possible interest for the encoding of quantum information. (We have cut off the figure at $\lambda/r=10$, where r is the nearest-neighbor distance, in order to retain the visibility of some of the oscillations at low values of the argument, which correspond to shorter wavelengths.)

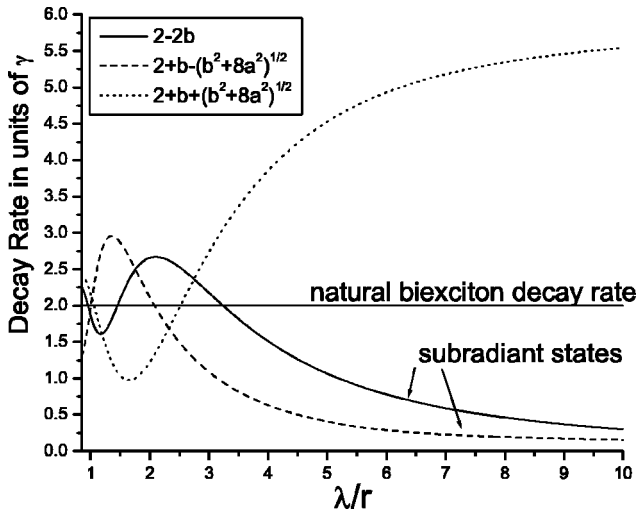

FIG. 2. Distance dependence of the decay rates of the $N=4$, $n=2$ eigenstates.

C. Triple excitation eigenstates

To complete the sets of eigenstates for the $N=6$ and $N=7$ polygons, we require those corresponding to the $n=3$ excitations. These are obtained with methods very similar to those used for the $n=2$ states. For $N=7$, the $M^{(3)}$ matrix is first partitioned into a 5×5 array of square submatrices, each of dimension 7×7. The solution then requires the (preliminary) diagonalization of a 5×5 matrix $M(P)$, but is otherwise a direct extension of the method used for $n=2$. For $N=6$, we choose the basis vectors in the order: $\{|B_{124}\rangle,|B_{235}\rangle,\ldots,|B_{613}\rangle;|B_{134}\rangle,|B_{245}\rangle,\ldots,|B_{623}\rangle;|B_{135}\rangle,|B_{246}\rangle\}$. Doing so we find that the matrix $M^{(3)}$ consists of a core array of 3×3 submatrices, each of dimension 6×6, together with an outer section of 2 columns to the right and 2 rows at the bottom of the core. The solution consists of 2 stages: In the first stage, the eigenvalues and eigenvectors of the core matrix are found, and in the second the symmetric and antisymmetric eigenvectors of the core are extended to become those of the full 20×20 matrix, in a manner entirely analogous to that employed for $N=6$, $n=2$. The complete set of eigenvalues and eigenvectors for $N=6$ is listed in Table III. Those for $N=7$ are available upon request.

II. OTHER STRUCTURES

We have calculated the eigenstates of a number of other simple structures as well (such as short linear chains, parallelograms, trapezoids, etc.), using symmetry considerations and brute force; results for these are available upon request. In this section, we present a few examples of (3-dimensional) structures for which the energy levels and eigenstates can be calculated by adapting the methods developed in Sec. I above.

A. "Diamond" structures

We consider a system consisting of an N-sided polygon of TLA's, together with one TLA above, and one below the center of the polygon. The $n=1$ eigenstates of this system

can be obtained by the same method as that used in Sec. I B for the calculation of the $n=2$ eigenstates of the $N=4$ polygon: The interaction matrix consists of an $N\times N$ ''core'' which is the interaction matrix $M^{(1)}$ of the polygon, augmented by an outer section of 2 extra columns to the right and rows at the bottom of the core. For example, the interaction matrix for a ''diamond'' based on an $N=6$ polygon is

$$M=\begin{pmatrix} 0 & a & b & c & b & a & d & e \\ a & 0 & a & b & c & b & d & e \\ b & a & 0 & a & b & c & d & e \\ c & b & a & 0 & a & b & d & e \\ b & c & b & a & 0 & a & d & e \\ a & b & c & b & a & 0 & d & e \\ d & d & d & d & d & d & 0 & f \\ e & e & e & e & e & e & f & 0 \end{pmatrix},$$

where $\Omega_{i7}\equiv d$ for $i=1,\ldots,6$, $\Omega_{i8}\equiv e$, and $\Omega_{78}\equiv f$.

The first 5 eigenvectors and eigenvalues of this matrix are those listed in Table III, corresponding to the $n=1$, $v=1,...,5$ states of the hexagon, each appended by two 0's. The remaining 3 eigenvectors are of the form $u=(1,1,1,1,1,1;x,y)$ and correspond to eigenvalues m such that x, y and m satisfy the equations

$$m-dx-ey=2a+2b+c, \quad mx-fy=6d, \quad my-fx=6e.$$

For a symmetric diamond, $e=d$ and $y=x$ and there remain only 2 such symmetric eigenstates; the third is replaced by the antisymmetric vector $u=(0,0,0,0,0,0;1,-1)$ and corresponds to eigenvalue $m=-f$. (For a ''pyramid,'' $e=f=y=0$, and there remain 2 symmetric eigenstates.)

In the long wavelength limit, the antisymmetric state and the states corresponding to $v=1,...,5$ are all optically inactive, while the 2 symmetric $v=6$ states are (in general) active. For example, for a structure having atoms 7 and 8 equally spaced above and below the hexagon center by a distance equal to the nearest-neighbor separation in the hexagon, the following values result:

$$m_1=(2.64V+4.423i)\gamma, \quad x_1=-0.367+0.0024i,$$

$$m_2=(-0.38V+3.577i)\gamma, \quad x_2=8.123+0.053i.$$

Here, $\hbar V\gamma$ is the nearest-neighbor interaction energy in the hexagon.

B. Polygon multilayers

The method introduced in Sec. I B can also be used to calculate the $n=1$ eigenstates of structures consisting of l layers of N-sided polygons, arrayed in parallel planes and centered on the same axis. The polygons need not be of the same linear dimension, and/or may be rotated relative to each other by an arbitrary angle (although aligned vertically or rotated by angles which are integer multiples of π/N give the simplest solutions). The interaction matrix consists of an $l\times l$ array of square submatrices, each of dimension $N\times N$. Each submatrix has as its eigenvectors those of the N-sided polygon, and those of the full matrix are obtained by first diagonalizing the $l\times l$ auxiliary matrix and then proceeding as in Sec. I B.

For example, the interaction matrix for 2 layers of pentagons, either aligned vertically or with a relative rotation angle of $\pi/5$, has the form

$$M(P)=\begin{pmatrix} M_{11}(P) & M_{12}(P) \\ M_{21}(P) & M_{22}(P) \end{pmatrix},$$

where the submatrices are given by $M_{11}(P)=a(P+P^4)+b(P^2+P^3)$, $M_{12}(P)=M_{21}(P)=cI+d(P+P^4)+e(P^2+P^3)$, and $M_{22}(P)=\alpha(P+P^4)+\beta(P^2+P^3)$. The eigenvectors of each submatrix are the vectors $u_{(v)}$ of the pentagon listed in Table I for $n=1$, $v=1,...,5$; their eigenvalues are $m_{11}(v)=2ac_{2v}+2bc_{4v}$, $m_{12}(v)=m_{21}(v)=c+2dc_{2v}+2ec_{4v}$, and $m_{22}(v)=2\alpha c_{2v}+2\beta c_{4v}$, where $c_j\equiv\cos(j\pi/5)$. Proceeding as in Sec. I B, we find for the eigenvalues of $M(P)$ the results

$$m_{\pm}(v)=\frac{1}{2}[m_{11}(v)+m_{22}(v)\pm Q(v)],$$

where

$$Q^2(v)=4[(a-\alpha)c_{2v}+(b-\beta)c_{4v}]^2+4[c+2dc_{2v}+2ec_{4v}]^2.$$

The associated eigenvectors are $U^{\pm}(v)=(V_1^{\pm}(v)u_{(v)};V_2^{\pm}(v)u_{(v)})$, where $V_1^{\pm}(v)=m_{11}(v)+m_{22}(v)\pm Q(v)$ and $V_2^{\pm}(v)=2m_{12}(v)$.

C. Icosahedron

The icosahedron consists of 2 pentagon layers of equal size, rotated relative to each other by an angle of $\pi/5$, with 2 additional atoms (symmetrically) placed on the symmetry axis above and below the center of the double layer at a height such that the nearest neighbor distances between all pairs of atoms are equal. Its eigenstates can be obtained with a combination of the methods in Secs. II A and II B above.

The double layer of pentagons has $\alpha=a$ and $\beta=b$. Its eigenvalues are therefore given by

$$m_{\pm}(v)=2ac_{2v}+2bc_{4v}\pm(c+2dc_{2v}+2ec_{4v}), \qquad (2)$$

and eigenvectors by $U^{\pm}(v)=(u_{(v)};\pm u_{(v)})$. The first 8 eigenvalues of the icosahedron are then given by $m_{\pm}(v)$ [Eq. (2)], and the associated eigenvectors by $W^{\pm}(v)=[U^{\pm}(v);0,0]$, $v=1,...,4$.

We denote by $f\equiv\Omega_{i\,11}\equiv\Omega_{j\,12}$, $g\equiv\Omega_{j\,11}\equiv\Omega_{i\,12}$ and $h\equiv\Omega_{11\,12}$, $i=1,...,5$ and $j=6,...,10$. Two more eigenvectors are then $W^{+}(5)=(1,1,1,1,1;1,1,1,1,1;x,x)$ and correspond to eigenvalues μ such that x and μ are given by

$$\mu_{\pm}=\frac{1}{2}[m_{+}(5)+h\pm R], \quad 2(f+g)x_{\pm}=h-m_{+}(5)\pm R,$$

where $R^2\equiv(h-m_{+}(5))^2+20(f+g)^2$. The last 2 eigenvectors are of the form

TABLE III. Eigenvalues and eigenvectors of 6 TLA's arranged on the vertices of a regular hexagon.

n	v	Eigenvalues	Eigenvectors
1	1,5	$a-b-c$	$(1,-1,-2,-1,1,2);\ (1,1,0,-1,-1,0)$
	2,4	$-a-b+c$	$(-1,-1,2,-1,-1,2);\ (1,-1,0,1,-1,0)$
	3	$-2a+2b-c$	$(-1,1,-1,1,-1,1)$
	6	$2a+2b+c$	$(1,1,1,1,1,1)$
2	1,5	$\pm\sqrt{b^2+3a^2}$	$(1,-1,-2,-1,1,2;\ 0,-u_\pm,-u_\pm,0,u_\pm,u_\pm;\ 0,0,0)$
			$(3,3,0,-3,-3,0;\ 2u_\pm,u_\pm,-u_\pm,-2u_\pm-u_\pm,u_\pm;\ 0,0,0)$
	2,4	ν	$(-1,-1,2,-1,-1,2;\ -2x,x,x,-2x,x,x;\ -y,2y,-y)$
			$(1,-1,0,1,-1,0;\ 0,-x,x,0,-x,x;\ -y,0,y)$
	3	$-2b$	$(-1,1,-1,1,-1,1;\ 0,0,0,0,0,0;\ 0,0,0)$
		$2b$	$(0,0,0,0,0,0;\ -1,1,-1,1,-1,1;\ 0,0,0)$
	6	μ	$(1,1,1,1,1,1;\ r,r,r,r,r,r;\ s,s,s)$
		$\nu^3+2b\nu^2+(4ac-b^2-3a^2-4c^2)\nu+2b(b^2-a^2+4ac)=0$	
		$x=(\nu^2+b\nu-2b^2)/[\nu(a-2c)-2ab]$	
		$\mu^3-4b\mu^2-4(2ac+b^2+3a^2+c^2)\mu+16b(b^2-a^2-2ac)=0$	
		$y=2(a\nu+2bc)/[\nu(a-2c)-2ab]$	
		$r=(\mu^2-2b\mu-8b^2)/2[\mu(a+c)+4ab]$	
		$s=2(a\mu+2bc)/[\mu(a+c)+4ab]$	
		$u_\pm=-b\pm\sqrt{b^2+3a^2}$	
3	1,5	$a-b-c$	$(m_\pm,-m_\pm,-2m_\pm,-m_\pm,m_\pm,2m_\pm;\ 1,-1,-2,-1,1,2;\ -1,-2,-1,1,2,1;\ 0,0)$
		$+(a+b)m_\pm$	$(m_\pm,m_\pm,0,-m_\pm,-m_\pm,0;\ 1,1,0,-1,-1,0;\ 1,0,-1,-1,0,1;\ 0,0)$
	1,5	$b\mp a\mp c$	$(0,0,0,0,0,0;\ -2,\mp1,1,\pm2,1,\mp1;\ \pm1,-1,\mp2,-1,\pm1,2;\ 0,0)$
	2,4		
			$(0,0,0,0,0,0;\ 0,-1,\mp1,0,\pm1,1;\ 1,\pm1,0,\mp1,-1,0;\ 0,0)$
	2,4	$c-a-b$	$(n_\pm,n_\pm,-2n_\pm,n_\pm,n_\pm,-2n_\pm;\ 1,1,-2,1,1,-2;\ 1,-2,1,1,-2,1;\ 0,0)$
		$+(a-b)n_\pm$	$(n_\pm,-n_\pm,0,n_\pm,-n_\pm,0;\ 1,-1,0,1-1,0;\ -1,0,1,-1,0,1;\ 0,0)$
	3	σ_-	$(-p_-,p_-,-p_-,p_-,-p_-,p_-;\ 1,-1,1,-1,1,-1;\ -1,1,-1,1,-1,1;\ -q_-,q_-)$
	3	$\mp c\pm2a-2b$	$(0,0,0,0,0,0;\ \mp1,1,\mp1,1,\mp1,1;\ -1,\pm1,-1,\pm1,-1,\pm1;\ 0,0)$
	6		
	6	σ_+	$(p_+,p_+,p_+,p_+,p_+,p_+;\ 1,1,1,1,1,1;\ 1,1,1,1,1,1;\ q_+,q_+)$
		$\sigma_\pm^3-2b\pm3c\pm2a)\sigma_\pm^2-[4(a^2+b^2)+(2a\pm2b-c)^2]\sigma_\pm\pm3c[c^2+2(\pm bc\mp4ab+ac)]=0$	
		$p_\pm=2[(2b\pm a)\sigma_\pm+3ca]/[\sigma_\pm^2\mp2c\sigma_\pm-3c^2]$	
		$q_\pm=6[a\sigma_\pm\mp ac+2bc]/[\sigma_\pm^2\mp2c\sigma_\pm-3c^2]$	
		$m_\pm=[2c+b-a\pm\sqrt{(2c+b-a)^2+8(a+b)^2}]/[2(a+b)]$	
		$n_\pm=[a+b-2c\pm\sqrt{(2c-b-a)^2+8(a-b)^2}]/[2(a-b)]$	

$$W^-(5)=(1,1,1,1,1;-1,-1,-1,-1,-1;y,-y)$$

and correspond to eigenvalues ν such that y and ν are given by

$$\nu_\pm=\frac{1}{2}[m_-(5)-h\pm S],\quad 2(f-g)y_\pm=-(h+m_-(5))\pm S, \tag{3}$$

where $S^2\equiv(h+m_-(5))^2+20(f-g)^2$. In the long wavelength limit, only the states $W^+(5)$ are optically active, with eigenvalues and vectors given by

$$\mu_+=(0.00002V+11.98882i)\gamma,\quad x_+=0.99874+0.08190i,$$

$$\mu_-=(-0.02137V+0.01118i)\gamma,$$

$$x_-=-4.97288+0.40782i.$$

D. Body-centered icosahedron

The eigenstates of an icosahedron with one more atom added at its body center are very similar. There are 8 eigenvalues $m_\pm(v)$ corresponding to $v=1,...,4$, with the associated eigenvectors $(U^\pm(v);0,0,0)$. Two more eigenvectors are given by $(1,1,1,1,1;-1,-1,-1,-1,-1;y,-y,0)$, with eigenvalues $\nu_\pm$ of Eq. (3). Finally, the last 3 eigenvectors are of the form $(1,1,1,1,1;1,1,1,1,1;x,x,y)$ and correspond to eigenvalues μ such that x, y and μ satisfy the equations

$$\mu-(f+g)x-jy=m_+(5),$$

$$(\mu-h)x-ky=5(f+g),$$

$$-2kx+\mu y=10j,$$

where $j\equiv\Omega_{i\,13}$, $i=1,...,10$, and $k\equiv\Omega_{11\,13}\equiv\Omega_{12\,13}$. In the long wavelength limit, these last 3 states are optically active, with eigenvalues and vectors given by

$$\mu_1=(-1.55290V+12.95930i)\,\gamma,$$

$$x_1=1.03235+0.12245i,\quad y_1=0.94949-0.09023i,$$

$$\mu_2=(3.34074V+0.03909i)\,\gamma,$$

$$x_2=-11.47272+4.74688i,$$

$$y_2=16.19381-5.82416i,$$

$$\mu_3=(-3.80918V+0.00161i)\,\gamma,$$

$$x_3=-2.76006+0.08068i,$$

$$y_3=-4.51950+0.10698i.$$

E. Dodecahedron

The dodecahedron can be considered as consisting of 4 layers of pentagons, of which those in layers 1 (top) and 4 (bottom) are of equal size but smaller than those in layers 2 and 3. Layers 1 and 2 are aligned with each other, as are layers 3 and 4; however, the top pair is rotated relative to the bottom by an angle of $\pi/5$. The layers are sized and spaced such that the distances between all pairs of nearest neighbors are equal.

The interaction matrix consists of a 4×4 array of submatrices, each of dimension 5×5. Using the method of Sec. II B, we first find the eigenvalues and vectors of the auxiliary matrix M,

$$M=\begin{pmatrix} A & C & D & F\\ C & B & E & D\\ D & E & B & C\\ F & D & C & A\end{pmatrix}.$$

Here, A is the 5×5 submatrix representing the interactions between the atoms within layer 1 (and 4); B represents the interactions between the atoms within layer 2 (and 3); C represents those between the atoms of layers 1 and 2 (and 3 and 4), D those between the atoms of layers 1 and 3 (and 2 and 4), E those between atoms of layers 2 and 3, and F those between atoms of layers 1 and 4. The eigenvalues of M are given by

$$\mu_\pm=\frac{1}{2}(A+B+E+F\pm R)$$

corresponding to the eigenvectors

$$U^\pm=\begin{pmatrix} A-B-E+F\pm R\\ 2(C+D)\\ 2(C+D)\\ A-B-E+F\pm R\end{pmatrix},$$

and

$$\nu_\pm=\frac{1}{2}(A+B-E-F\pm S)$$

corresponding to the eigenvectors

$$V^\pm=\begin{pmatrix} A-B+E-F\pm S\\ 2(C-D)\\ -2(C-D)\\ -(A-B+E-F\pm S)\end{pmatrix},$$

where $R^2=(B-A-E+F)^2+4(C-D)^2$ and $S^2=(B-A+E-F)^2+4(C-D)^2$.

The eigenvalues and vectors of the dodecahedron are then found by substituting the eigenvalues $a(v)$ for A, $b(v)$ for B, etc.; they are given by

$$\mu_\pm(v)=\frac{1}{2}[a(v)+b(v)+e(v)+f(v)\pm R(v)]$$

corresponding to the eigenvectors

$$W^\pm(v)=(U_1^\pm(v)u_{(v)};U_2^\pm(v)u_{(v)};U_3^\pm(v)u_{(v)};U_4^\pm(v)u_{(v)})$$

and

$$\nu_\pm(v)=\frac{1}{2}[a(v)+b(v)-e(v)-f(v)\pm S(v)]$$

corresponding to the eigenvectors

$$Z^\pm(v)=(V_1^\pm(v)u_{(v)};V_2^\pm(v)u_{(v)};V_3^\pm(v)u_{(v)};V_4^\pm(v)u_{(v)}),$$

where $u_{(v)}$ are the eigenvectors of the pentagon and $v=1,...,5$. In the long-wavelength limit, only the states $W^\pm(5)$ are optically active, with eigenvalues and eigenvectors given by

$$\mu_+=(3.05984V+10i)\,\gamma,$$

$$U_1^+=U_4^+=0.5588+0.6906i/V,$$

$$U_2^+=U_3^+=-0.4396+2.2882i/V,$$

$$\mu_-=(0.04101V+10i)\,\gamma,$$

$$U_1^-=U_4^-=0.4005+0.8775i/V5,$$

$$U_2^-=U_3^-=0.5827-2.4829i/V.$$

III. CONCLUSIONS

We have calculated the eigenstates corresponding to coherent single and multiple excitations of an array of N identical TLA's arranged on the vertices of a regular polygon, and of a number of related 3-dimensional structures. The atomic coupling occurs via an exchange interaction which depends only on their separation. We include the interactions between all pairs, and our results are valid for arbitrary distances relative to the radiation wavelength. These states are used in [8] to study the absorptive and emissive properties of a number of polygon systems.

ACKNOWLEDGMENTS

We wish to thank Tamlyn Rudolph who performed some of the preliminary calculations, and Roger Mong for assistance with the dodecahedron. One of us (H.F.) gratefully acknowledges the hospitality of the Department of Physics of the University of Toronto, where this work was completed. This research was supported in part by the Natural Sciences and Engineering Research Council of Canada, the Austrian Science Foundation FWF, the TMR programs of the European Union, Project No. ERBFMRXCT960087, and by the NSA & ARO under Contract No. DAAG55-98-C-0040.

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