Cooperative Ordering in Lattices of Interacting Dipoles

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Using classical electrodynamics simulations we investigate the cooperative behavior of regular monolayers
of induced two-level dipoles, including their cooperative decays and shifts. For the particular case of the
kagome lattice we observe behavior akin to EIT for lattice spacings less than the probe wavelength. Within this region
the dipoles exhibit ferroelectric and anti-ferroelectric ordering. We also model how the cooperative response is
manifested in the optical transmission through the kagome lattice, with sharp changes in transmission from 10% to
80% for small changes in lattice spacing.

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If dipoles in an ensemble radiate coherently then the overall response of the ensemble is dramatically altered, e.g. when all
the emitters are well within a wavelength of each other [1, 2] or equally if the emitters are regularly spaced [3–5]. One such example of ‘cooperative’ [6] behavior is a shortening (super-radiance) or lengthening (subradiance) of the excitation lifetime of the ensemble. This is often accompanied by an energy shift and there are many examples of these cooperative decays and shifts in both theory [1, 3–5, 7–9] and experiment [10–14]. This, along with other exotic phenomena such as highly directional scattering [15] and strong excitation localization [16–18], has the potential to produce a vast range of applications, from long lived quantum memories to simulation of complex phenomena (e.g. excitation hopping [18, 19]).

Cooperative effects have been studied for regular arrays and random ensembles in 1D [3, 5, 9, 18], 2D [5, 16, 17, 20] and 3D [15, 20, 21], however the particular focus of our paper is the case of interacting dipolar spins in geometries with constrained dynamics. One such example is the 2D kagome lattice which has been shown in different spin systems to demonstrate spin-ice and geometric frustration [22], photonic flat bands and band gaps [23, 24] as well as low loss transmission through hollow-core photonic crystal fibers [25, 26]. We will show how the kagome lattice now also exhibits interesting cooperative behavior. Such a kagome spin lattice could be implemented for example in cold atoms [27–33], paramagnetic molecular chains [34] and microfabricated plasmonic nanostructures [35].

The configurations we consider in this letter are regular 2D arrays in the xy plane of N identical discrete driven dipoles. We will often relate this to the case of 2-level atoms with a J = 0 → J = 1 electric dipole transition (e.g. as in Sr [36]) although much of the physics is general to other kinds of dipoles. In contrast to [16] we do not restrict the dipole moments to any particular direction, which for atoms would mean the excited Zeeman sub-levels are degenerate. The magnitude and direction of the ith dipole moment d(i) (i ∈ 1,...,N), located at position r(i), is proportional to the classical electric field incident on it E(r(i)) (for details of this model see [20, 37–39]):

\[
d(i) = \alpha E(r(i)),
\]

where we take the constant of proportionality to be the 2-level polarizability \( \alpha = -D^2/(\hbar(\Delta + i\Gamma_0/2)) \). This in turn depends on the dipole moment matrix element D, the detuning of the electric field from the resonance frequency \( \Delta \), and the FWHM transition linewidth \( \Gamma_0 = D^2k_0^2/3\pi\hbar\epsilon_0 \) (where \( k_0 \) is the wavenumber corresponding to the resonant wavelength \( \lambda_0 = 2\pi/k_0 \)). Each dipole radiates a secondary electric field; hence the total field \( E \) felt by a dipole located at position \( r(j) \) is the sum of the applied driving field \( E_0 \) and the fields \( E^{(j)} \) radiated from all other dipoles:

\[
E(r(j)) = E_0(r(j)) + \sum_{j\neq i} E^{(j)}(r(j)).
\]

The field radiated by dipole j is proportional to its dipole moment through \( E^{(j)}(r) = G(r - r^{(j)})d^{(j)} \), where G is the dipole propagation tensor (as given in [40]). The matrix elements in a Cartesian representation of G are given by

\[
G_{p,q}(r) = \frac{1}{\epsilon_0} \left\{ \frac{\partial}{\partial r_p} \frac{\partial}{\partial r_q} - \delta_{p,q} \nabla^2 \right\} \frac{\epsilon_0 r}{4\pi r^3} - \delta_{p,q} \delta(r),
\]

where \( p, q \in \{1, 2, 3\} \). \( r_1, r_2, r_3 \) are the components of \( r \) directed along the \( \{\hat{x}, \hat{y}, \hat{z}\} \) unit vectors, and \( r = |r| \). Substituting (2) into (1) yields a linear system of coupled equations,

\[
d^{(j)} = \alpha E_0(r^{(j)}) + \alpha \sum_{j\neq i} G(r - r^{(j)})d^{(j)},
\]

which, to reiterate, describes how the dipole moment of dipole \( i \) is proportional to the applied external field (first term on right) as well as the field radiated from every other dipole (second term on right). (4) can be solved self-consistently [20] to calculate \( d^{(j)} \) for an ensemble of dipoles in any arbitrary arrangement. In this letter we consider exclusively the driving light to be propagating along \( z \), and linearly polarized in y (\( E_0 = \hat{y} \)).

Since \( E_0 \) is real and the polarizability \( \alpha \) is in general complex, each dipole moment \( d^{(j)} \) contains in general a real part which corresponds to the component of the dipole excitation that is in-phase with the driving field, and an imaginary part which is the out-of phase component. For example, a single independent dipole driven on resonance by \( E_0 \) will be purely imaginary (\( \pi/2 \) out of phase with the driving field).
Before modeling many-atom lattices it is instructive to consider the basic building blocks or unit cells that make up the larger lattices. Using (4) we calculate what the steady-state dipole moments are for four minimal configurations of between \( N = 3 \) and \( N = 7 \) dipoles corresponding to square, triangular, hexagonal, and kagome (trihexagonal) lattice geometries. The configurations we use aren’t necessarily the true ‘unit cell’ or ‘basis’ of the lattice but do have some respective connections to the aforementioned lattices. In Fig. 1 we plot the mean dipole moment amplitude, \( \bar{d} = \frac{\sum_{i=1}^{N} \left| \mathbf{d}^{(i)} \right|}{N} \), which gives us a measure of how strongly on average the dipoles are responding to the driving field.

Our model assumes weak driving [20, 38] which for the case of atoms would mean excited state populations can be neglected. In Fig. 1 each panel shows a strong broad feature at large spacings which corresponds to the state where the dipoles oscillate in phase [18]. This state gets weaker at smaller spacings allowing us to see other narrower resonances due to some of the other \( N - 1 \) normal modes (this is highlighted by the cross-section cut in 1(a), see e.g. [2]). Whilst here we have plotted the average dipole amplitude, we get qualitatively similar plots for just the imaginary dipole moments (as considered later in Fig. 3), which is akin to the susceptibility of a continuous polarizable medium and so a broadening or narrowing of the width with respect to the natural linewidth corresponds to a shortening or lengthening of the excitation lifetime. This is called superradiance and subradiance respectively [41].

Already for these simple configurations the kagome (bow-tie) base unit [Fig. 1(d)] is beginning to display qualitatively different resonance features, with more, stronger lines and a strong resonance at red detunings. These differences become more striking as we increase the number of dipoles.

In Fig. 2 we plot average dipole moment amplitude for \( N = 49 \) dipoles in a square lattice (c) and \( N = 47 \) dipoles in a kagome lattice (d). We consider a Gaussian driving beam with waist \( w = 2\lambda \) [42]. Again we see lots of narrow subradiant resonances and one broad (alternating superradiant and subradiant) resonance which dominates at large spacings. What we notice in the kagome case however which we don’t see in the square (nor the triangular and hexagonal lattices which aren’t shown here) is a striking region close to resonance in which there is weak dipolar response surrounded on either side of detuning by regions of strong dipolar response (highlighted by the white dashed ring). We shall now examine this more closely.

So far we have just considered the average dipole mag-
nitude which gives us no information about the direction or complex nature of the dipolar response. In Fig. 3(a) and 3(b) we now plot the real and imaginary parts of the \( y \) component of the dipole moments, \( \text{Re}\[ \bar{d}_y \] and \( \text{Im}\[ \bar{d}_y \] \) respectively, again averaging over all the dipoles in each lattice. For a real driving field linearly polarized in \( y \), \( \text{Re}\[ \bar{d}_y \] \) and \( \text{Im}\[ \bar{d}_y \] \) are related respectively to the dispersion and decay due to the dipoles by the optical theorem [43]. The shift and FWHM of \( \text{Im}\[ \bar{d}_y \] \) calculated for a pair of atoms exactly agrees with the analytic shift and width of the two atom decay derived in [9].

In Fig. 3(c) we plot the cross-section of \( \text{Re}\[ \bar{d}_y \] \) and \( \text{Im}\[ \bar{d}_y \] \) through the region of interest we highlighted in Fig. 2(d) as a function of detuning and observe a striking resemblance to the absorptive and dispersive lineshapes through an electromagnetically induced transparency (EIT) resonance.

In EIT quantum interference between excitation pathways connecting different energy states leads to a transparency window on resonance in which the pathways interfere destructively and a probe field is allowed to pass through the scattering medium unattenuated. In our dipolar ensemble we have a set of \( N \) different cooperative states or normal modes. Some are broad and others narrow, and as they scan over each other they have the potential to interfere in a similar way. For most of the weak narrow states passing over the strong broad state we see Fano-like asymmetric resonances. However this transparency region is much stronger than these resonances (one of which can be seen faintly as a bump on the solid purple line around \( \Delta = 0.3\Gamma_0 \) in Fig. 3(c)) and is similar in appearance to a broad EIT transparency.

Other similar coupled systems have been shown to demonstrate EIT-like transparency [44–48]. However, as in [47], our transparency region is due to interplay between cooperative resonances of the entire ensemble (when e.g. considering an ensemble of driven atoms) as opposed to conventional EIT where transparency comes from interference between resonances within a single scatterer.

The question remains however; what is so special about the kagome lattice that it allows for this transparency region when the other lattices do not? As mentioned earlier kagome spin lattices are associated with spin-ice and geometric frustration and so looking at what the individual dipole moments themselves are doing may give a clue. In Fig. 3(d,e,f) we plot the individual dipole moments in the \( xy \) plane (the \( z \) components are zero due to the system geometry) for several atoms at the center of a kagome lattice with \( N = 355 \). This bigger lattice behaves qualitatively similar to the smaller \( N = 47 \) lattice but the different dipole behaviors are more distinct at large \( N \).

In the blue-detuned feature (Fig. 3(d) - circle) the imaginary part is dominant and the dipoles tend to align with the driving field along \( y \). This is the same as the limit of large separation where interactions are dominated by the driving. We refer to this behavior as ‘ferroelectric’. Moving to the transparency window (Fig. 3(e) - star) the real part is now dominant and the dipoles inhabit the mode in which the angle between nearest neighbors is minimized. In the kagome geometry this is satisfied with nearest neighbors oriented at 120° to each other.

We refer to this behavior ‘anti-ferroelectric’. Finally we move past the transparency window to the red-detuned strong feature (Fig. 3(f) - triangle) where again the response is predominantly out of phase with the driving field. This is now more of a composite state in which dipoles on one third of the lattice sites align strongly with the field whilst the remaining dipoles strongly anti-align with each other along the \( x \) direction.

It is intriguing that the region of transparency in the kagome
lattice coincides with strong dispersive and anti-ferroelectric behavior in the dipoles. This particular 120° anti-ferroelectric configuration cannot be realized in the same way in the other lattice geometries which may help to explain why it is only in the kagome lattice that we observe such a region of transparency.

Whilst this remains to be investigated further, it is clear however that there exists a wealth of rich cooperative behavior in these kagome and other lattices which can be explored not just directly through observation of the dipole moments themselves but also through indirect measurements such as the optical transmission and extinction through the lattices.

In Fig. 4 we calculate the resonant transmission of the driving field as a function of lattice spacing through a $N = 165$ kagome lattice. We do this by integrating the total electric field as a function of lattice spacing through a $N$ itself but also through indirect measurements such as the optical transmission and extinction through the lattices. In conclusion, we have shown that dipoles arranged in a regular 2D lattice respond to driving light cooperatively rather than independently and this response depends on lattice geometry, lattice spacing and detuning of the driving field. We observe cooperative decays and shifts akin to those predicted for pairs [9] and 1D chains [18] of atomic dipoles, with different superradiant and subradiant cooperative states being populated. Unique to the kagome lattice however is a region of transparency through which the driven dipole moment is less than for nearby parameters. The in-phase and out-of-phase components of the average dipole moments appear similar to the absorption and dispersion of an EIT-like resonance respectively. Within this transparency region the dipoles align anti-ferroelectrically. The cooperative response of the dipoles also affects the optical transmission through the dipolar lattice. In future studies we hope to investigate further the spatial scattering pattern around the lattices as well as go beyond steady-state classical models to include dynamics, e.g. excitation hopping.

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\begin{equation}
T = \frac{\int |E|^2 dS}{\int |E_0|^2 dS},
\end{equation}

where $E$ is calculated by replacing $r^{(i)}$ with $r$ in (2). As observed in [20] there is a strong deviation from the non-interacting random ensemble limit given by the Beer-Lambert decay (purple dashed line). However because of the periodicity in the lattice there are strong resonances above and below this non-interacting limit followed by a complete transparency again at very small spacing corresponding to when the dipoles are shifted completely off-resonance. Now the region we called the region of ‘transparency’ appears as a peak in the transmission at around $a = 0.4\lambda$ and around either side of this peak as well as at $a = \lambda$ the transmission varies very rapidly as a function of spacing. These steep variations would hopefully be possible to observe in an experiment by small changes in the lattice spacing and may even provide a mechanism towards an application such as a highly sensitive pressure switch. In future work we will investigate the effect of lattice imperfections on these cooperative features and the challenge experimentally becomes how to create a tunable trapping potential with sub-wavelength lattice spacings and limit dissipation.

The inset shows the total electric field intensity as the driving field propagates through the same kagome lattice at a spacing of $a = 0.3\lambda$. It is clear to see the extinction of the light downstream of the dipoles (in the +$z$ direction) and the corresponding strong back reflection.

\[ \text{FIG. 4. (Color online) Calculated optical transmission through a kagome lattice of } N = 165 \text{ dipoles driven by a Gaussian beam with waist } w = 2\lambda \text{ and linearly polarized in } y. \text{ The black solid line plots the transmission for the full scattering model (5). The purple dashed line plots the transmission for a random ensemble of non-interacting dipoles with an equivalent uniform 2D number density } \rho_{2D} \text{ and optical cross-section } \sigma \text{ calculated using the Beer-Lambert exponential decay, } T = \exp(-\sigma r_{2D}). \text{ The inset shows extinction and backward reflection of the total field through the kagome lattice with spacing } a = 0.3\lambda \text{ as indicated by the arrow. The small green dots are dipole positions and black dashed lines show the } 1/e \text{ width of the driving beam.} \]
to highlight the multiple-scattering nature of our dipoles as opposed to ‘collective’ first order mean field behavior. This is not however a universal convention.

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