

Laser field control of subradiant states of a system of several atoms in a configuration close to a regular polygon

A.A. Makarov, V.I. Yudson

Abstract. Some collective excitations of an ensemble of closely spaced (two-level) atoms can be characterised by extremely low rates of radiation decay, which is of considerable interest for storing quantum information. Such (dark) states are realised, for example, in planar geometry when atoms are located at the nodes of a regular polygon. However, the decay rate of such excitations increases markedly when the geometry deviates from ideal. Using the example of small deformation of a square in a rhombus, we show that an increase in the decay rate of the dark state due to symmetry breaking can be compensated for by adjusting the transition frequency of some atoms exposed to external laser radiation.

Keywords: atomic ensembles, spontaneous decay, subradiative states, quantum information.

1. Introduction

A system of several identical atoms enclosed in a volume with a linear size $r < \lambda_{eg}$ (λ_{eg} is the wavelength of the transition from the excited state $|e\rangle$ of a single atom to the ground state $|g\rangle$) was first considered by Dicke [1] 65 years ago as an example of the formation of superradiant and subradiant states. Singly excited states Q_n of the system consisting of N noninteracting atoms can be expressed in general form as

$$Q_n = |g_1 \dots g_{n-1} e_n g_{n+1} \dots g_N\rangle, \quad (1)$$

where the atom with the number n is excited, and the remaining atoms are in the ground state. The dipole–dipole interaction between them leads to the formation of N superpositional (entangled) states, the expected properties of which can correspond either to the so-called *superradiance* (spontaneous decay that is faster than the $|e\rangle \rightarrow |g\rangle$ transition in a single atom) or to *subradiance* (slow spontaneous decay). This classification is readily realised at $N = 2$. In this case, the dipole–dipole interaction of the Q_1 and Q_2 states leads to the formation of two entangled eigenstates: symmetric, $Q_s = 2^{-1/2}(Q_1 + Q_2)$, and antisymmetric, $Q_a = 2^{-1/2}(Q_1 - Q_2)$ [2, 3].

While the Q_s state at $r \ll \lambda_{eg}$ decays at a rate $\Gamma_s \approx 2\Gamma$ (that is, with a double rate of the spontaneous decay of a single atom), for the Q_a state we have $\Gamma_a \sim (r/\lambda_{eg})^2\Gamma$. When studying this system, the main attention in a number of works (see, for example, [4–6] and review [3]) was focused on superradiance. An interesting effect of switching between the Q_1 and Q_2 states, caused by the variation of the spatial structure of the exciting laser pulse, was considered by Das et al. [7]. As for the subradiant states, an increased interest in them was caused by their slow spontaneous decay and the resulting potential for long-term storage of quantum information.

In connection with this, the logical question is whether the subradiant states of atomic configurations whose decay rate in the parameter r/λ_{eg} is lower than in the above case are possible. A striking example of this kind was proposed by V.S. Letokhov [8–10]: two atoms in the 1D case (one-dimensional waveguide or photonic crystal) located at a distance equal to an integer number of half-waves, $m\lambda_{eg}/2$. The antisymmetric state with an even value of m and the symmetric state with an odd value of m are superstable; they are sustained by a field filling the space between the atoms. The decay of such states with a small difference in the distance from its critical value was studied by Redchenko and Yudson [11]. In a one-dimensional waveguide, more complex configurations were also considered (see, for example, [12]). However, such schemes can only be realised for spatially separated atoms. For two closely spaced atoms in the three-dimensional (3D) case, we note two schemes for controlling subradiant states [13, 14]. An interesting (although rather complicated) scheme for constructing a single-quantum subradiant state in an ensemble of a large number of atoms was considered by Scully [15], and the possibility of its control was discussed by Cai et al. [16]. As for the experiment, some evidence in favour of a change in the spontaneous decay rate was obtained, for example, by Pavolini et al. [17] for an ensemble of a large number of atoms and by DeVoe and Brewer [18] for a system of two ions in a trap. Finally, more recently, subradiance from a cloud of cold atoms was reliably observed [19]. The contribution of this effect was small, but detectable by a narrow spectrum of fluorescence delayed by the times significantly exceeding the time of superradiance.

In connection with the foregoing, of fundamental importance is the formation of subradiant states that decay more slowly with respect to the r/λ_{eg} parameter than in a system of two atoms [for example, as $(r/\lambda_{eg})^4$, etc.]. More recently, the ‘theorem of existence’ of such states was proved [20, 21]. This is a system that demonstrates the effect of extraordinary subradiance, i.e. ‘quantum emitters’ (atoms?) in the configuration (geometry) of a regular polygon, starting with a square (number of atoms $N \geq 4$).

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In this work, we study modifications of the extraordinary subradiance effect with respect to small changes in the optimal geometry. It turns out that changes in shape in the particular case $N = 4$ (from square to rhombus) can be fully compensated for by shifting the transition frequency in two opposite atoms. In Section 2, we formulate the model and present general analytical results that make it possible to calculate the rates of spontaneous single-quantum decay of atomic systems in arbitrary geometry. Section 3 discusses the properties of excited single-quantum eigenstates of atomic systems in the geometry of regular polygons. In Section 4, we present the results of calculating the spontaneous decay rate of the lower state of the system in rhombic geometry with finding the frequency shifts that are optimal for these structures. Finally, in Section 5, we propose a scheme using laser radiation, which allows one to realise the necessary energy shifts.

2. Spontaneous emission of atomic systems

We consider a system of N identical atoms localised at the points $\mathbf{R}_n = (x_n, y_n, z_n = 0)$ on the plane $z = 0$. For simplicity, we assume that the ground state $|g\rangle$ of the atom is nondegenerate (the total angular momentum $J_g = 0$), and the excited state $|e\rangle$ is three-fold degenerate ($J_e = 1$) with projections $M = 0, \pm 1$ on the z axis (for example, atoms of group II, i.e. Be, Mg, Ca, etc.). We restrict ourselves to the states with the projection $M = 0$, i.e., linearly polarised radiation with the transition $|e^{(0)}\rangle \rightarrow |g\rangle$. The Hamiltonian of the subsystem of singly excited states has the form:

$$\hat{H} = \begin{pmatrix} E & U_{12} & \dots & U_{1n} & \dots & U_{1N} \\ U_{21} & E & \dots & U_{2n} & \dots & U_{2N} \\ \dots & \dots & \dots & \dots & \dots & \dots \\ U_{n1} & U_{n2} & \dots & E & \dots & U_{nN} \\ \dots & \dots & \dots & \dots & \dots & \dots \\ U_{N1} & U_{N2} & \dots & U_{Nn} & \dots & E \end{pmatrix}, \quad (2)$$

where E is the energy of the excited state $|e\rangle$ of a single atom, which we further consider equal to zero; and U_{jk} are the matrix elements of the dipole–dipole interaction operator, which depend on the distance $r_{jk} = \sqrt{(x_j - x_k)^2 + (y_j - y_k)^2}$ between atoms for a given pair of them. In the case under study, when the quantisation axis of the angular momentum projection is perpendicular to the line connecting the atoms, in the expression for the dipole–dipole interaction operator

$$\hat{U} = \frac{(\hat{\mathbf{d}}_j \cdot \hat{\mathbf{d}}_k)}{r^3} - 3 \frac{(\hat{\mathbf{d}}_j \cdot \mathbf{r})(\hat{\mathbf{d}}_k \cdot \mathbf{r})}{r^5} \quad (3)$$

only the first term remains. Therefore, $U_{jk} = |\langle e^{(0)} | \hat{\mathbf{d}}_z | g \rangle|^2 \times r_{jk}^{-3}$, and it is convenient to express these matrix elements in terms of the spontaneous emission rate Γ on the transition $|e^{(0)}\rangle \rightarrow |g\rangle$:

$$\Gamma = \frac{32\pi^3}{3\hbar\lambda_{\text{eg}}^3} |\langle e^{(0)} | \hat{\mathbf{d}}^{(z)} | g \rangle|^2. \quad (4)$$

Then,

$$U_{jk} = \frac{3\Gamma}{4(kr_{jk})^3}, \quad (5)$$

where $k = 2\pi/\lambda_{\text{eg}}$. Next, we will characterise the system of atoms using the parameter U_r , which is equal to the value of the *smallest* matrix element U_{jk} corresponding to the *maximum* distance between the atoms in the system.

Having formulated assumptions and limitations, we present a formula for the spontaneous emission rates of an arbitrary ‘planar’ system of atoms from eigenstates $|\psi_j\rangle$ ($j = 1, 2, \dots, N$) constructed taking into account the dipole–dipole interaction as a superposition of states (1). The derivation of the general formula is given in Appendix 1. In the particular case of plane geometry and the transition without changing the angular momentum projection onto an axis perpendicular to the plane $z = 0$, the spontaneous emission rate from the state $|\psi_j\rangle$ is expressed in terms of the coordinates \mathbf{R}_n of the atoms and the projections C_{nj} of states (1) onto the state $|\psi_j\rangle$ as

$$\Gamma_j^{(N)} = \frac{3\Gamma}{8\pi} \int_0^{2\pi} \int_0^\pi \left| \sum_{n=1}^N C_{nj} \exp(i\mathbf{k}\mathbf{R}_n) \right|^2 \sin^3\theta d\theta d\varphi. \quad (6)$$

Here, the components of the vector \mathbf{k} are related to the spherical angles θ and φ by ordinary expressions: $\mathbf{k} = k(\sin\theta \cos\varphi, \sin\theta \sin\varphi, \cos\theta)$. Formula (6) is used in further calculations.

3. Regular polygons of atoms as a source of extraordinary subradiant states

Let us consider the change in the structure of singly excited energy states with an increase in the number of atoms in the configuration of a regular polygon. In the trivial case of $N = 2$, the energies $E_1 = -U_r$, $E_2 = +U_r$, while the eigenvectors are antisymmetric and symmetric, respectively, since the interaction is a positive quantity. The atomic coordinates are selected in the form $\mathbf{R}_1 = \|-r/2, 0, 0\|$, $\mathbf{R}_2 = \|+r/2, 0, 0\|$. The decay rates of the states $|1\rangle$ and $|2\rangle$ in accordance with (6) are expressed as:

$$\Gamma_1^{(2)} = \frac{3\Gamma}{8\pi} \int_0^{2\pi} \int_0^\pi \sin^2\left(\frac{1}{2}kr \sin\theta \cos\varphi\right) \sin^3\theta d\theta d\varphi, \quad (7)$$

$$\Gamma_2^{(2)} = \frac{3\Gamma}{4\pi} \int_0^{2\pi} \int_0^\pi \cos^2\left(\frac{1}{2}kr \sin\theta \cos\varphi\right) \sin^3\theta d\theta d\varphi,$$

where the subscript at Γ denotes the state number, and the upper one is the number of atoms. When $kr \ll 1$, we have

$$\Gamma_1^{(2)} \approx \frac{3}{16}(kr)^2 \Gamma \int_0^\pi \sin^5\theta d\theta = \frac{1}{5}(kr)^2 \Gamma, \quad \Gamma_2^{(2)} \approx 2\Gamma. \quad (8)$$

In the case of $N = 3$ (an equilateral triangle), nothing fundamentally changes. There are two levels: a doubly degenerate level with an energy $E_1 = E_2 = -U_r$, the decay rate of which at $kr \ll 1$ is proportional to $(kr)^2$, and a level with an energy $E_3 = 2U_r$, which exhibits a triple decay rate. However, at $N = 4$ (a square), we observe a new qualitative effect in comparison with the previous two cases. The three upper levels form a structure similar to that at $N = 3$: the upper state with $E_4 = (4\sqrt{2} + 1)U_r$ decays at $(kr)^2 \ll 1$ at a rate of 4Γ , the next level is twice degenerate ($E_{2,3} = -U_r$) and its decay rate is proportional to $(kr)^2$. A significant difference is that the rate of spontaneous emission from the nondegenerate lower state [its energy is $E_1 = (-4\sqrt{2} + 1)U_r$, and the eigenvector is $C_{n1} = (1/2)(-1)^n$] is already proportional to $(kr)^4$ at $kr \ll 1$; in the general case, according to (6), we have




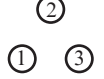


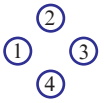



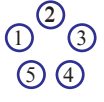



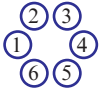



Geometry	Eigenstates	Decay rates Γ_i
	 Symmetric  Asymmetric	$\Gamma_i \approx 2\Gamma$ $\propto (r/\lambda)^2$
	 Symmetric  Asymmetric	$\Gamma_i \approx 3\Gamma$ $\propto (r/\lambda)^2$
	 Symmetric  Asymmetric  Asymmetric	$\Gamma_i \approx 4\Gamma$ $\propto (r/\lambda)^2$ $\propto (r/\lambda)^4$
	 Symmetric  Asymmetric  Asymmetric	$\Gamma_i \approx 5\Gamma$ $\propto (r/\lambda)^2$ $\propto (r/\lambda)^4$
	 Symmetric  Asymmetric  Asymmetric	$\Gamma_i \approx 6\Gamma$ $\propto (r/\lambda)^2$ $\propto (r/\lambda)^4$ $\propto (r/\lambda)^6$

Figure 1. Atomic systems in the geometry of regular polygons, their eigenstates and the corresponding spontaneous decay rates.

$$\begin{aligned}
 \Gamma_1^{(4)} &= \frac{3\Gamma}{8\pi} \int_0^{2\pi} \int_0^\pi \left[\cos\left(\frac{1}{2}kr \sin\theta \sin\varphi\right) \right. \\
 &\quad \left. - \cos\left(\frac{1}{2}kr \sin\theta \cos\varphi\right) \right]^2 \sin^3\theta d\theta d\varphi \\
 &= \frac{3\Gamma}{2\pi} \int_0^{2\pi} \int_0^\pi \sin^2\left[\frac{1}{4}kr \sin\theta(\cos\varphi + \sin\varphi)\right] \\
 &\quad \times \sin^2\left[\frac{1}{4}kr \sin\theta(\cos\varphi - \sin\varphi)\right] \sin^3\theta d\theta d\varphi. \quad (9)
 \end{aligned}$$

Then, at $kr \ll 1$ we find

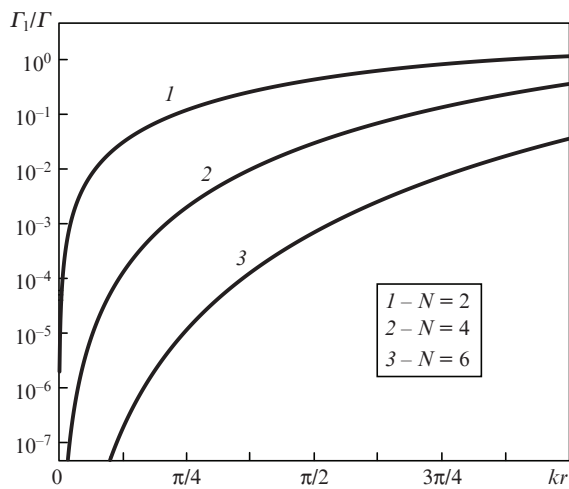


Figure 2. Dependences of the spontaneous decay rates of the lower state of systems of two, four, and six atoms on the size of the atomic system with the geometry shown in Fig. 1.

$$\begin{aligned}
 \Gamma_1^{(4)} &\approx \frac{3}{512\pi} (kr)^4 \Gamma \int_0^{2\pi} \int_0^\pi \sin^7\theta \cos^2 2\varphi d\theta d\varphi = \\
 &= \frac{3}{512} (kr)^4 \Gamma \int_0^\pi \sin^7\theta d\theta = \frac{3}{560} (kr)^4 \Gamma \approx 5.4 \times 10^{-3} (kr)^4 \Gamma. \quad (10)
 \end{aligned}$$

A similar pattern is established with a further increase in the number of atoms (Fig. 1). Thus, at $N = 5$, one more state is added to the lower state, so that the lower level becomes doubly degenerate, and then at $N = 6$ a new nondegenerate level is added below, the decay rate of which at $kr \ll 1$ is proportional to $(kr)^6$. Figure 2 shows a series of dependences of $\Gamma_1^{(N)}$ on the system size r .

4. System of four atoms in the geometry of a rhombus: effect of a shift in the energy levels of one of the opposite atomic pairs

The lifetime of extraordinary subradiant states turns out to be very sensitive to small changes in geometry. This circumstance is illustrated in Fig. 3, which shows the dependence of the decay rate of the lower state on the angle characterising the deviation of the rhombic geometry from the square geometry. One can see that when $kr \approx 1$, Γ increases by 2–3 times in the transition from a square (with an increase in the obtuse angle by about 10°). However, at $kr \ll 1$, the growth rate of decay Γ_1 increases by orders of magnitude.

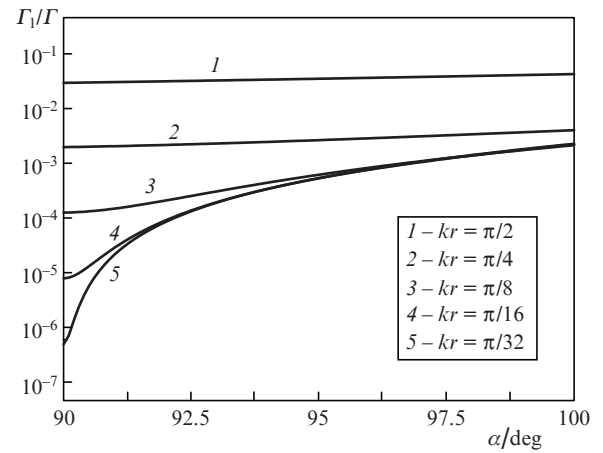


Figure 3. Dependences of the spontaneous decay rate of the lower state of a system of atoms in the geometry of a rhombus on an obtuse angle at two opposite vertices, presented for different distances kr between atoms located at vertices at an acute angle.

Thus, with a relatively small change in structure, the ‘extraordinary subradiance’ effect is radically destroyed. Its restoration (and even some enhancement) is possible by shifting the transition frequency in a pair of atoms located at opposite vertices of the rhombus relative to the transition frequency in another pair of atoms. In particular, for atoms located at the vertices of an obtuse angle, the shift $\Delta\omega$ must be negative. The dependences of Γ_1 on $\Delta\omega$, shown in Fig. 4, demonstrate sharp minima at optimal values of the frequency shift. The minimum values of Γ_1 for the corresponding optimal $\Delta\omega$ are given in Table 1 for various r . Analytical expressions for the eigenstates and energies of the rhombic configuration of atoms are given in Appendix 2.

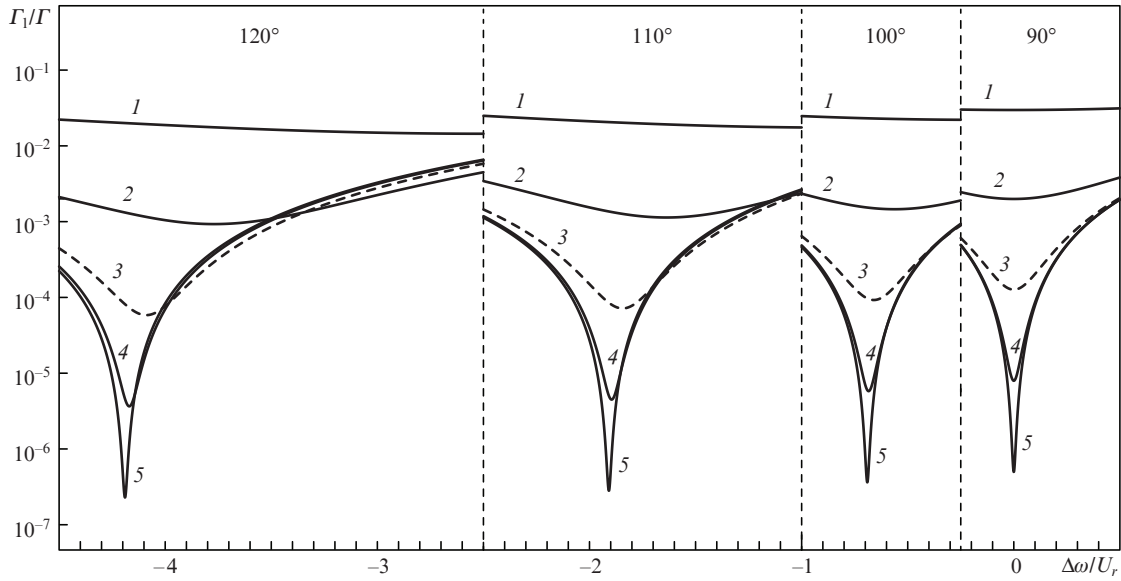


Figure 4. Dependences of the spontaneous decay rate of the lower state of the atomic system in the geometry of a rhombus on the transition frequency shift in a pair of atoms at obtuse vertices for different distances in another pair of atoms at $kr = (1) \pi/2$, $(2) \pi/4$, $(3) \pi/8$, $(4) \pi/16$ and $(5) \pi/32$.

Table 1. The optimal energy shift of two atoms located at ‘obtuse’ vertices of the rhombus, and the corresponding decay rates of an extraordinary subradiant state.

$(kr)^2$	$\alpha = 90^\circ$		$\alpha = 100^\circ$		$\alpha = 110^\circ$		$\alpha = 120^\circ$	
	$\Delta\omega/U_r$	Γ_1/Γ	$\Delta\omega/U_r$	Γ_1/Γ	$\Delta\omega/U_r$	Γ_1/Γ	$\Delta\omega/U_r$	Γ_1/Γ
0.01	0	5.4×10^{-7}	-0.69	3.9×10^{-7}	-1.91	3.0×10^{-7}	-4.19	2.5×10^{-7}
0.10	0	5.3×10^{-5}	-0.67	3.9×10^{-5}	-1.87	3.0×10^{-5}	-4.13	2.5×10^{-5}
1.00	0	5.2×10^{-3}	-0.48	3.8×10^{-3}	-1.47	3.0×10^{-3}	-3.51	2.4×10^{-3}

5. Inducing a shift by a laser field in the form of a standing wave

Of the available scenarios of a frequency shift in two pairs of atoms (in obtuse- and acute-angle vertices of a rhombus), we consider the use of a standing laser wave (Fig. 5). It is possible, for example, as shown in the figure, to use a transition from the ground state $|g\rangle$ into an auxiliary state $|a\rangle$ located higher in energy than the state $|e\rangle$. This scheme can be of

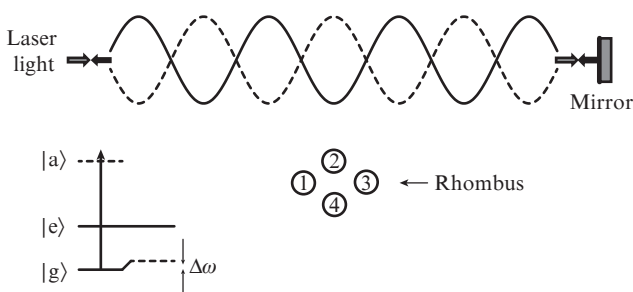


Figure 5. Scheme for inducing an energy shift of the ground state in a pair of atoms located in the obtuse vertices of a rhombus. The laser frequency is detuned relative to the frequency of the transition from the ground state to the high-lying excited state of the atom to the blue. The antinodes of a standing wave are at the position of a pair of atoms 2–4, the nodes are at the position of a pair of atoms 1–3.

purely practical value provided that at $kr \lesssim 1$ the laser wavelength that is close to the transition wavelength $|g\rangle \rightleftharpoons |a\rangle$ will be able to provide the necessary excess of the amplitude of the field acting on a pair of atoms 2–4 over the amplitude of the field acting on a pair of atoms 1–3. We should also note the possibility of varying the field difference due to the inclination of the standing wave with respect to the plane in which the atoms reside. In any case, the difference in transition frequencies $|e\rangle \rightarrow |g\rangle$ in this scenario is expressed in terms of the difference in energy shifts

$$\Delta\omega \approx \frac{(d_{ga}\mathcal{E})^2}{4\hbar^2(\omega_{las} - \omega_{ag})} \quad (11)$$

of the ground state of two pairs of atoms due to the difference in the squares of the amplitudes of the standing wave field \mathcal{E} ; here, d_{ga} is the matrix element of the dipole moment operator for the transition $|g\rangle \rightleftharpoons |a\rangle$.

6. Conclusions

We have analysed the properties of subradiant states decaying more slowly than $(kr)^2\Gamma$ (see Fig. 2) for a system of closely spaced ($r/\lambda \ll 1$) atoms in the geometry of a regular polygon. We have shown that, with a relatively small violation of the geometry, the spontaneous decay rate of these states increases dramatically (see Fig. 3). However (and this is the main result of the work), the situation is corrected due to a shift in the transition frequency in certain atoms with respect to other

atoms. The calculations have been performed only for the geometry of a rhombus (see Fig. 4 and Table 1), but it seems that higher-order polygons should also exhibit the observed effects. They are especially pronounced when the distance between the atoms is much less than the wavelength of the transition in use. We can note that the problem of *subwavelength localisation* has been developed recently in a number of theoretical works. Thus, one of the latest original proposals is based on high-frequency modulation of the optical field [22]. In addition to laser tweezers and ion traps, the introduction to [22] reviews other scenarios considered in the literature. For example, Dubetsky and Berman [23] consider the use of optical lattices based on multiphoton optical transitions to be a promising method. We can also note the idea of a femtosecond trap (see, for example, [24]). The considered trend, as a prelude to new interesting physics, is topical and will be unconditionally developed. Solving the subwavelength localisation problem will allow one to use the ideas of this work for ultra-precise measurements of the characteristics of systems of closely spaced atoms. The measurable quantity in the experiment does not have to be the decay rate; apparently, it is more convenient to diagnose subradiant states by measuring the radiation linewidth.

It should be noted separately a scenario when the frequency shift does not need to be induced, i.e. when it is natural, for example, in isotopes. Then the control is reduced to solving the inverse problem, i.e. manipulation of the geometry (with the fine tuning of the laser radiation) to minimise the rate of spontaneous emission of a certain superpositional (entangled) state.

Appendix 1

We obtain a formula for the spontaneous decay rate of an arbitrary singly excited eigenstate of a system of N atoms, which is represented as an expansion in many-particle basis states similar to (1), without restricting ourselves to considering plane geometry and sampling of excited states with only zero angular momentum projection M :

$$|\Psi\rangle = \sum_{M=-1}^1 (C_1^{(M)} |e_1^M g_2, \dots, g_N\rangle + C_2^{(M)} |g_1 e_2^{(M)}, \dots, g_N\rangle + \dots + C_N^{(M)} |g_1, \dots, g_{n-1}, \dots, e_n^{(M)}\rangle) \equiv \sum_{i=1}^N \sum_{m=-1}^1 C_i^{(M)} \psi_i^{(M)}, \quad (\text{A1.1})$$

where for definiteness we consider the same particular case $J_g = 0$ and $J_e = 1$, as in the main text. The interaction of each atom with a plane wave is described [25, 26] as

$$\hat{\mathcal{H}}' = \frac{\sqrt{3\Gamma}}{4\pi |\langle e^0 | \hat{\mathbf{d}} | g \rangle|} (\hat{\mathbf{d}} \cdot \mathbf{e}_{k\rho}) \exp(i\mathbf{k}\mathbf{R}) \equiv \frac{\sqrt{3\Gamma}}{4\pi} (\hat{\mathcal{O}}_1 + \hat{\mathcal{O}}_2). \quad (\text{A1.2})$$

Here, $\mathbf{e}_{k\rho}$ is a pair of orthogonal unit vectors ($\rho = 1, 2$) perpendicular to the wave vector \mathbf{k} ; \mathbf{R} is the radius vector of the position of the atom; and the subscripts at $\hat{\mathcal{O}}$ correspond to the photon polarisation ρ . It is convenient to choose the vectors $\mathbf{e}_{k\rho}$ in the form, corresponding to expression of the vector \mathbf{k} in polar coordinates $\mathbf{k}/|\mathbf{k}| = (\sin\theta \cos\varphi, \sin\theta \sin\varphi, \cos\theta)$:

$$\begin{aligned} \mathbf{e}_{k1} &= (-\sin\varphi, \cos\varphi, 0), \\ \mathbf{e}_{k2} &= (\cos\theta \cos\varphi, \cos\theta \sin\varphi, -\sin\theta). \end{aligned} \quad (\text{A1.3})$$

Then we find the scalar products $\hat{\mathbf{d}} \cdot \mathbf{e}_{k\rho}$ included in formula (A1.2):

$$(\hat{\mathbf{d}} \cdot \mathbf{e}_{k1}) = -\frac{i}{2} \hat{d}_+ \exp(-i\varphi) + \frac{i}{2} \hat{d}_- \exp(i\varphi), \quad (\text{A1.4})$$

$$(\hat{\mathbf{d}} \cdot \mathbf{e}_{k2}) = \frac{1}{2} \hat{d}_+ \cos\theta \exp(-i\varphi) + \frac{1}{2} \hat{d}_- \cos\theta \exp(i\varphi) - \hat{d}_z \sin\theta.$$

Next, we write out the matrix elements of transitions from the singly excited states of the i th atom:

$$\begin{aligned} \langle \psi_i^{-1} | \hat{\mathcal{O}}_1 | g_1, \dots, g_N \rangle &= \frac{i}{\sqrt{2}} \exp(i\varphi) \exp(i\mathbf{k}\mathbf{R}_i), \\ \langle \psi_i^0 | \hat{\mathcal{O}}_1 | g_1, \dots, g_N \rangle &= 0, \\ \langle \psi_i^+ | \hat{\mathcal{O}}_1 | g_1, \dots, g_N \rangle &= -\frac{i}{\sqrt{2}} \exp(-i\varphi) \exp(i\mathbf{k}\mathbf{R}_i), \\ \langle \psi_i^{-1} | \hat{\mathcal{O}}_2 | g_1, \dots, g_N \rangle &= \frac{1}{\sqrt{2}} \cos\theta \exp(i\varphi) \exp(i\mathbf{k}\mathbf{R}_i), \\ \langle \psi_i^0 | \hat{\mathcal{O}}_2 | g_1, \dots, g_N \rangle &= -\sin\theta \exp(i\mathbf{k}\mathbf{R}_i), \\ \langle \psi_i^+ | \hat{\mathcal{O}}_2 | g_1, \dots, g_N \rangle &= \frac{1}{\sqrt{2}} \cos\theta \exp(-i\varphi) \exp(i\mathbf{k}\mathbf{R}_i). \end{aligned} \quad (\text{A1.5})$$

Finally, we express through these matrix elements the spontaneous decay rate of the state (A1.1):

$$\begin{aligned} \tilde{\Gamma} &= \frac{3\Gamma}{8\pi} \int_0^{2\pi} \int_0^\pi \left(\left| \sum_{i=1}^N \sum_{m=-1}^1 C_i^m \langle \psi_i^m | \hat{\mathcal{O}}_1 | g_1, \dots, g_N \rangle \right|^2 \right. \\ &\quad \left. + \left| \sum_{i=1}^N \sum_{m=-1}^1 C_i^m \langle \psi_i^m | \hat{\mathcal{O}}_2 | g_1, \dots, g_N \rangle \right|^2 \right) \sin\theta d\theta d\varphi. \end{aligned} \quad (\text{A1.6})$$

Here the factor in front of the integral is obtained from the golden rule in the form of $2\pi(\sqrt{3\Gamma}/4\pi)^2$.

Next, we can perform a numerical calculation for any configuration of atoms and any orientation of the axes. In the case of plane geometry and taking into account only components with $M = 0$, the terms with the matrix elements of the operator $\hat{\mathcal{O}}_1$ vanish and formula (A1.6) is reduced to formula (6) in Section 2.

Appendix 2

We label the vertices of the rhombus sequentially with numbers from 1 to 4, assuming that the internal angle α at vertices 2 and 4 is obtuse. Let the distance between vertices 1 and 3 be equal to r . We denote the matrix element of the interaction between these vertices by $U_{13} = U$ (in the text, by definition, $U_{13} = U_r$; hereinafter, we omit the subscript r). Then the remaining elements of the interaction matrix have the form:

$$\begin{aligned} U_{12} &= U_{23} = U_{34} = U_{14} = 8U \sin^3(\alpha/2), \\ U_{24} &= U \tan^3(\alpha/2). \end{aligned} \quad (\text{A2.1})$$

The frequencies of opposite atoms 2 and 4 are assumed to be shifted by $\Delta\omega$ with respect to the frequencies of atoms 1 and 3. The system of equations for the amplitudes C_j of the eigenvector $|\psi\rangle$ has the form

$$\begin{aligned} EC_1 &= U_{12}(C_2 + C_4) + UC_3, \\ EC_2 &= \Delta\omega C_2 + U_{12}(C_1 + C_3) + U_{24}C_4, \\ EC_3 &= U_{12}(C_2 + C_4) + UC_1, \\ EC_4 &= \Delta\omega C_4 + U_{12}(C_1 + C_3) + U_{24}C_2. \end{aligned} \quad (\text{A2.2})$$

It is convenient to introduce new variables:

$$\begin{aligned} A &= \frac{1}{2}(C_1 + C_2), \quad a = \frac{1}{2}(C_1 - C_2), \\ B &= \frac{1}{2}(C_2 + C_4), \quad b = \frac{1}{2}(C_2 - C_4). \end{aligned} \quad (\text{A2.3})$$

Then the system of equations (A2.2) reduces to the form

$$\begin{aligned} (E + U)a &= 0, \\ (E - \Delta\omega + U_{24})b &= 0, \\ (E - U)A - 2U_{12}B &= 0, \\ -2U_{12}A + (E - \Delta\omega - U_{24})B &= 0. \end{aligned} \quad (\text{A2.4})$$

The four eigenenergies are given by the expressions*

$$\begin{aligned} E_a &= -U, \\ E_b &= \Delta\omega - U_{24} = \Delta\omega - U \tan^3(\alpha/2), \end{aligned}$$

$$\begin{aligned} E_{\pm} &= \frac{1}{2}(U + U_{24} + \Delta\omega) \pm \sqrt{\frac{1}{4}(U - U_{24} + \Delta\omega)^2 + 4U_{12}^2} \quad (\text{A2.5}) \\ &= U \left\{ \frac{1}{2} \left[1 + \tan^3(\alpha/2) + \frac{\Delta\omega}{U} \right] \right. \\ &\quad \left. \pm \sqrt{\frac{1}{4} \left[1 - \tan^3(\alpha/2) + \frac{\Delta\omega}{U} \right]^2 + 256 \sin^6(\alpha/2)} \right\}. \end{aligned}$$

In the case of a general position, when all the eigenenergies are different, the eigenvectors are determined by system (A2.4). For example, let $E = E_a$ be different from other eigenenergies. Then A , B and b vanish. For the amplitudes C_j , we find: $C_1 = -C_3 = 1/\sqrt{2}$ and $C_2 = C_4 = 0$. Adjusting the value of $\Delta\omega$ allows one to manipulate the eigenenergies and eigenstates so as to ensure a minimum decay rate (see Fig. 4 and Table 1).

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*For the square ($\alpha = 90^\circ$ and $\Delta\omega = 0$) there is a correspondence $E_{(a,b)} \Leftrightarrow E_{(2,3)}, E_+ \Leftrightarrow E_4, E_- \Leftrightarrow E_1$, if we number the energies in ascending order. However, in the general case, such a correspondence may be violated.