Entanglement on macroscopic scales in a resonant-laser-field-excited atomic ensemble

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We show that two groups of slow two-level atoms in a weak resonant laser field are entangled. The considered groups can be separated by a macroscopic distance, and be parts of a larger atomic ensemble. In a dilute regime, for two very distant groups of atoms, in a plane-wave laser beam, we determine the maximum attainable entanglement negativity, and a laser intensity below which they are certainly entangled. They both decrease with increasing distance between the two groups, but increase with enlarging groups sizes. As a consequence, for given laser intensity, far separated groups of atoms are necessarily entangled if they are big enough.

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I. INTRODUCTION

The impact of the environment on quantum entanglement is manifold. On the one hand, an initial entanglement between independent systems can be destroyed by their coupling to their surroundings. This fragility of quantum entanglement is substantiated by the fact that it can vanish in finite time, as first shown for two-level atoms in distinct vacuum cavities [1]. On the other hand, the environment mediates interactions, Hamiltonian or not, between the considered systems, and can then induce correlations between them, and hence potentially entanglement. It has been shown that finite entanglement can develop between two initially uncorrelated two-level systems, or qubits, sharing the same surroundings, but otherwise uncoupled [2–6]. However, in the realistic case of a finite separation between the two systems, this effect is only transient if the environment is in thermal equilibrium [5,6].

This is not the case when the surroundings does not reduce to a thermal bath. The systems steady state, reached asymptotically from any initial state, can present finite entanglement, as has been shown for two qubits in diverse environments, such as the electromagnetic vacuum and a resonant laser field [7,8], two heat reservoirs at different temperatures [9], and the electromagnetic field emitted by two bodies at different temperatures [10,11]. The entanglement obtained in these works can be essentially traced back to one of the two familiar features of a pair of infinitely close qubits, which are the decoupling, from the environment, of the so-called subradiant state, and the divergent energy shifts of the single-excitation levels [12–14]. Experimentally, transient entanglement of two atoms has been generated using the Rydberg blockade mechanism, which relies on similar dipole-dipole energy shifts, but of double-excitation levels [15,16]. Another approach consists of starting with multilevel atoms coupled to one or several monochromatic fields, and possibly to static fields or to a common cavity mode, and then reducing them to two-level systems by adiabatic elimination of upper levels in an appropriate regime [17–19]. Following the proposal of Ref. [18], long-lived entanglement of two atomic ensembles separated by 50 cm has been observed experimentally [20,21].

In this paper, we are concerned with the entanglement generated by a resonant laser field in an atomic ensemble. Entangled steady states of systems of many driven qubits have been obtained for highly simplified interaction models, in which each qubit is coupled identically to every other one [22], or is coupled only to its nearest neighbors in a chain [23]. We consider here two-level atoms evolving under the influence of a laser field and of the obviously present electromagnetic vacuum. Lehmberg's master equation is used to describe the dynamics of the atoms internal state [5,7,8,14,18,24]. For two atoms, the resulting steady state is separable or entangled, depending on the relative strength of the laser amplitude and vacuum-mediated interaction [7,8,22]. We focus on the regime of low laser intensities, in which entanglement is long range. As we will see, the underlying physical origin of the found entanglement is that, for weak laser fields, the atoms internal dynamics is dominated by the dipole-dipole interaction, laser photon absorption, and collective radiative decay, which all preserve the state purity. As a result, the atoms steady state is practically pure, and correlated, and hence entangled. As shown in the following, this remains true for large groups of atoms, and even if they are surrounded by other identical atoms, as illustrated in Fig. 1.

The rest of the paper is organized as follows. The Hamiltonian used to describe laser-excited two-level atoms, and the approximations leading to Lehmberg's master equation, are presented in the next section. In Sec. III, the steady internal state of slow-moving atoms in a weak resonant laser field, is determined, and two of its features, which are of particular importance for entanglement, are discussed. In Sec. IV, the entanglement of any two subgroups of atoms is studied. It is shown that there is a laser intensity threshold, which depends on the considered atoms, below which the two subgroups are entangled. More quantitative results are derived, in a dilute regime, for macroscopically distant groups of atoms. Finally, in the last section, we summarize our results, and mention some questions raised by our study.

II. MASTER EQUATION FOR LASER-EXCITED TWO-LEVEL ATOMS

We consider an ensemble of two-level atoms evolving under the influence of a laser field. Within the dipolar approximation for the coupling to the electromagnetic field [12], and a semiclassical approximation for the motion of the atoms [25], the dynamics of the atoms internal state is governed by the

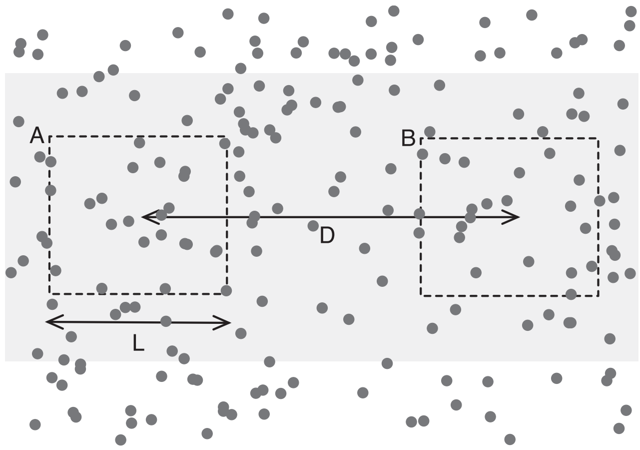

FIG. 1. Schematic representation of two subgroups of atoms, A and B, of characteristic size L, and separated by a distance D, of a larger atomic ensemble, partially illuminated by a resonant laser beam. A and B are entangled, for $D \simeq 1$ m and $L \simeq 50$ μm; for example, see Sec. IV C.

Hamiltonian

$$H = H_e + \omega_0 \sum_\mu \sigma_\mu^\dagger \sigma_\mu - \sum_\mu (\sigma_\mu + \sigma_\mu^\dagger) \times \left[\mathbf{d} \cdot \mathbf{E}(\mathbf{r}_\mu) + \frac{\Omega}{2} \mathrm{Re}[w(\mathbf{r}_\mu) e^{-i\omega t}] \right], \quad (1)$$

where ω_0 is the atomic resonance frequency, $\mathbf{r}_\mu$ is the classical position of atom μ, and Ω is proportional to the laser field amplitude. Throughout this paper, units are used in which $\hbar = 1$. The Hamiltonian H_e and field $\mathbf{E}$ read, respectively, $H_e = c \int d^3k |\mathbf{k}| (a_{\mathbf{k}1}^\dagger a_{\mathbf{k}1} + a_{\mathbf{k}2}^\dagger a_{\mathbf{k}2})$, and

$$\mathbf{E}(\mathbf{r}) = \int d^3k \left(\frac{-c|\mathbf{k}|}{16\pi^3 \epsilon_0} \right)^{1/2} \sum_{p=1,2} \mathbf{e}_{\mathbf{k}p} e^{i\mathbf{k}\cdot\mathbf{r}} a_{\mathbf{k}p} + \mathrm{H.c.}, \quad (2)$$

where c is the speed of light, ϵ_0 is the vacuum dielectric permittivity, $\mathbf{e}_{\mathbf{k}1}$ and $\mathbf{e}_{\mathbf{k}2}$ are unit vectors orthogonal to $\mathbf{k}$ and to each other, and the electromagnetic field operators $a_{\mathbf{k}p}$ satisfy the bosonic commutation relations $[a_{\mathbf{k}p}, a_{\mathbf{k}'p'}^\dagger] = \delta_{pp'} \delta(\mathbf{k} - \mathbf{k}')$. The atomic operator σ_μ is defined by $\sigma_\mu = |g\rangle_{\mu\mu}\langle e|$ where $|g\rangle_\mu$ and $|e\rangle_\mu$ are, respectively, the ground and excited states of atom μ. The vector $\mathbf{d} = {}_\mu\langle e|\mathbf{D}_\mu|g\rangle_\mu$ where $\mathbf{D}_\mu$ is the dipole moment of atom μ, is assumed real and the same for all the atoms. The spatial function w depends on the laser beam considered, $w(\mathbf{r}) = \exp(i\mathbf{K} \cdot \mathbf{r})$ for a plane wave of wave vector $\mathbf{K}$, for example.

For fixed positions $\mathbf{r}_\mu$, the time scales relevant to the dynamics of the atoms internal state ρ are ω_0^{-1}, $|\mathbf{r}_\mu - \mathbf{r}_\nu|/c$, Ω^{-1}, and Γ^{-1} where $\Gamma = |\mathbf{d}|^2 \omega_0^3 / 3\pi\epsilon_0 c^3$ is the spontaneous decay rate of an isolated atom [24,26,27]. In the following, we consider laser intensities such that $\Omega \ll \Gamma$. The ratio Γ/ω_0 is of the order of α^3 where $\alpha \simeq 7 \times 10^{-3}$ is the fine-structure constant [12]. Thus, for distances $|\mathbf{r}_\mu - \mathbf{r}_\nu| \ll k_0^{-1} \alpha^{-3}$ where $k_0 = \omega_0/c$, the time scale Γ^{-1} and Ω^{-1} are very long compared to the other ones, and it can be shown that the time evolution of ρ is well described by the master equation

$$\partial_t \rho = -\Gamma \mathcal{L}(\{\mathbf{r}_\mu\})\rho - i[H_a + H_l, \rho], \quad (3)$$

where H_a and H_l are, respectively, the second and last terms of Hamiltonian (1). The superoperator $\mathcal{L}$ is defined by

$$\mathcal{L}\varrho = \sum_{\mu,\nu} [z_{\mu\nu} \sigma_\mu^\dagger \sigma_\nu \varrho + z_{\mu\nu}^* \varrho \sigma_\mu^\dagger \sigma_\nu - 2\gamma_{\mu\nu} \sigma_\mu \varrho \sigma_\nu^\dagger], \quad (4)$$

where ϱ is any matrix, $\gamma_{\mu\nu} = \mathrm{Re} z_{\mu\nu}$, $z_{\mu\mu} = 1/2$, and, for $\mu \neq \nu$,

$$z_{\mu\nu} = \frac{3}{4} \frac{e^{ir}}{r^3} \{[1 - 3(\hat{\mathbf{d}} \cdot \hat{\mathbf{r}})^2](i + r) - i[1 - (\hat{\mathbf{d}} \cdot \hat{\mathbf{r}})^2] r^2\}, \quad (5)$$

with $r = k_0 |\mathbf{r}_\mu - \mathbf{r}_\nu|$, $\hat{\mathbf{r}} = (\mathbf{r}_\mu - \mathbf{r}_\nu)/|\mathbf{r}_\mu - \mathbf{r}_\nu|$, and $\hat{\mathbf{d}} = \mathbf{d}/|\mathbf{d}|$ [24,26]. For atoms moving with velocities $\partial_t \mathbf{r}_\mu \ll c/k_0 |\mathbf{r}_\nu - \mathbf{r}_\xi|$, Eq. (3) remains valid with the time-dependent positions $\mathbf{r}_\mu(t)$ [27]. Thus, for typical values of ω_0 and Γ, and velocities of the order of 10 m s^{-1}, Eq. (3) is relevant for interatomic distances as large as some decimeters.

III. INTERNAL STATE OF SLOW ATOMS IN A WEAK RESONANT LASER FIELD

We consider atoms' velocities such that

$$\partial_t \mathbf{r}_\mu \ll \Gamma k_0^{-1}. \quad (6)$$

In terms of the temperature T of the atomic ensemble, this condition can be rewritten as $T/A \ll 1$ K where A is the mass number of the atoms. Since the characteristic length scale of both H_l and $\mathcal{L}$, is k_0^{-1}, the displacements of the atoms during a time interval of length Γ^{-1}, can be neglected in Eq. (3). Consequently, at each instant t, ρ is essentially equal to the asymptotic solution of this equation with atoms' positions $\mathbf{r}_\mu(t)$ assumed fixed. It is hence of the form $\rho = \sum_p \rho_p \exp(-ip\omega t)$. Due to the small value of Γ/ω_0, the matrices ρ_p are practically given by their zeroth-order expansions in this ratio. Thus, they obey $[H_a, \rho_p] = p\omega_0 \rho_p$, and are determined by

$$i\mathcal{L}\rho_p + p\delta\rho_p + \eta[W, \rho_{p+1}] + \eta[W^\dagger, \rho_{p-1}] = 0, \quad (7)$$

where $\delta = (\omega - \omega_0)/\Gamma$ is a dimensionless laser detuning, which is assumed to be at most of order unity, $\eta = \Omega/2\Gamma$ and $W = \sum_\mu \sigma_\mu w(\mathbf{r}_\mu)^*$. Note that only the rotating wave part of H_l, i.e., $\Omega W \exp(i\omega t)/2 + \mathrm{H.c.}$, appears in these equations.

A. Low laser intensity perturbative solution

As we are concerned with laser intensities such that $\Omega \ll \Gamma$, we solve Eq. (7) perturbatively in the ratio η. To do so, we expand the Fourier components ρ_p as $\rho_p = \rho_p^{(0)} + \eta\rho_p^{(1)} + \cdots$, where $\rho_p^{(0)} = \delta_{p0} |\mathcal{G}\rangle\langle\mathcal{G}|$, with $|\mathcal{G}\rangle = \otimes_\mu |g\rangle_\mu$, is the solution to Eq. (7) for $\eta = 0$. From Eq. (7), the successive $\rho_p^{(n)}$ are related by

$$(i\mathcal{L} + p\delta)\rho_p^{(n+1)} = -\big[W, \rho_{p+1}^{(n)}\big] - \big[W^\dagger, \rho_{p-1}^{(n)}\big]. \quad (8)$$

Note that, strictly speaking, the regime of validity of the following results is $\alpha^3 \ll \eta \ll 1$, since, in the derivation of Eq. (7), terms of the order of $(\Gamma/\omega_0)^2$ have been neglected, whereas terms of the order of Ω/ω_0 have been kept.

Using the recursive relation (8), we find, up to second order in η,

$$\rho = (\langle\psi|\psi\rangle^{-1}|\psi\rangle\langle\psi|)^{[2]}, \tag{9}$$

where the superscript $^{[2]}$ means that only terms up to second order are kept, and

$$|\psi\rangle = |\mathcal{G}\rangle + \eta\sum_{\mu} u_\mu|\mu\rangle + \eta^2\sum_{\mu<\nu}(u_\mu u_\nu + v_{\mu\nu})|\mu\nu\rangle, \tag{10}$$

with $|\mu\rangle = \sigma_\mu^\dagger|\mathcal{G}\rangle$ and $|\mu\nu\rangle = \sigma_\mu^\dagger|\nu\rangle$; see Appendix A. The components u_μ and $v_{\mu\nu}$ obey

$$\begin{gathered}\sum_{\xi} z_{\mu\xi}u_\xi - i\delta u_\mu = i w_\mu,\\ \sum_{\xi}(z_{\mu\xi}\tilde{v}_{\xi\nu} + z_{\nu\xi}\tilde{v}_{\xi\mu}) - 2i\delta v_{\mu\nu} = z_{\mu\nu}\big(u_\mu^2 + u_\nu^2\big),\end{gathered} \tag{11}$$

where $\mu < \nu$, $w_\mu = w(\mathbf{r}_\mu)\exp(-i\omega t)$, $\tilde{v}_{\mu\nu}$ equals 0 for $\mu = \nu$, $v_{\mu\nu}$ for $\mu < \nu$, and $v_{\nu\mu}$ for $\mu > \nu$.

The state of atom μ, which reads $\rho_\mu = |\phi\rangle_{\mu\mu}\langle\phi| - \eta^2|u_\mu|^2|g\rangle_{\mu\mu}\langle g|$ with $|\phi\rangle_\mu = |g\rangle_\mu + \eta u_\mu|e\rangle_\mu$, is determined by u_μ, and the correlations between atoms μ and ν are determined by $v_{\mu\nu}$, since $\rho_{\mu\nu} - \rho_\mu\otimes\rho_\nu = \eta^2 v_{\mu\nu}^*\sigma_\mu\sigma_\nu + \text{H.c.}$ where $\rho_{\mu\nu}$ is the state of the pair of atoms μ and ν. Note that these correlations vanish if the mutual influence between the atoms, mediated by the electromagnetic vacuum, is neglected, since Eqs. (11) give $v_{\mu\nu} = 0$ for $z_{\mu\nu} = \delta_{\mu\nu}/2$. In the absence of vacuum-mediated interaction, entanglement between atoms can still be generated using photons, but not with a simple laser field [28–30].

B. Schrödinger-like equation

The fact that ρ coincides, up to second order, with a pure state plays an essential role in the following. The origin of this effective purity can be understood as follows. The first two terms of the superoperator (4), which describe the dipole-dipole interaction between the atoms and the decay of the excited atomic levels due to spontaneous emission, can be interpreted in terms of an effective complex Hamiltonian. This is not the case of the last one, which accounts for the populating, by spontaneous emission, of $\langle k|\rho|l\rangle$ where $|k\rangle$ and $|l\rangle$ are eigenstates of H_a, from matrix elements $\langle k'|\rho|l'\rangle$ such that $\epsilon_{k'} > \epsilon_k$ where $\epsilon_k = \langle k|H_a|k\rangle$. However, for small η, and in the long-time regime, this process essentially does not contribute to ρ, since the order, in η, of $\langle k|\rho|l\rangle$ increases with ϵ_k. Up to second order, it only results in a correction to the ground-state population $\langle\mathcal{G}|\rho|\mathcal{G}\rangle$, which simply ensures the normalization of ρ. Due to the decline of $\langle k|\rho|l\rangle$ with increasing ϵ_k, stimulated emission is also negligible. Consequently, Eq. (7) can be approximated by the Schrödinger-like equation

$$\partial_t|\psi\rangle = -\left[iH_a + \Gamma\sum_{\mu,\nu} z_{\mu\nu}\sigma_\mu^\dagger\sigma_\nu - i\frac{\Omega}{2}e^{-i\omega t}W^\dagger\right]|\psi\rangle, \tag{12}$$

where the last term describes laser photon absorption. This equation is satisfied, up to second order, by a state of the form (10), provided u_μ and $v_{\mu\nu}$ fulfill Eqs. (11).

C. State of a subensemble

An important property of the state (9) is that the ensuing state of any subensemble of atoms is given by an expression of the same form. Consider the system S consisting of the atoms $\mu = 1,\ldots,n$, and the complementary system $\overline{\text{S}}$ consisting of all the other atoms. The pure state (10) can be expanded on the basis $\{|\mathcal{G}\rangle_{\overline{\text{S}}},\sigma_\mu^\dagger|\mathcal{G}\rangle_{\overline{\text{S}}},\ldots\}$ of system $\overline{\text{S}}$, where $|\mathcal{G}\rangle_{\overline{\text{S}}} = \otimes_{\mu>n}|g\rangle_\mu$, as $|\psi\rangle = |\mathcal{G}\rangle_{\overline{\text{S}}}|\psi\rangle_{\text{S}} + \cdots$. The state $|\psi\rangle_{\text{S}}$ is given by expression (10), but with sums running only over the first n atoms, and $|\mathcal{G}\rangle$, $|\mu\rangle$, and $|\mu\nu\rangle$, replaced by the corresponding states for system S. The point is that the following terms in the above expansion of $|\psi\rangle$ either do not contribute to the second-order state ρ_{S} of system S, or contribute only to a correction to the population ${}_{\text{S}}\langle\mathcal{G}|\rho_{\text{S}}|\mathcal{G}\rangle_{\text{S}}$, which simply ensures the normalization of ρ_{S}. Consequently, ρ_{S} is given by Eq. (9) with $|\psi\rangle$ replaced by $|\psi\rangle_{\text{S}}$.

IV. ENTANGLEMENT

In this section, we discuss the entanglement of any two subgroups of atoms, say A and B, such as those schematically depicted in Fig. 1. A sufficient, but in general not necessary, condition for A and B to be entangled, is that the partial transpose ρ_{AB}^Γ of their collective state ρ_{AB} has negative eigenvalues. A resulting measure of the entanglement between A and B is the negativity $\mathcal{N}$, which is the absolute sum of the negative eigenvalues of ρ_{AB}^Γ. It vanishes for separable states, and is equal to 1/2 for two-qubit maximally entangled states [31]. To study the entanglement of A and B for low laser intensities, i.e., $\Omega \ll \Gamma$, we determine the first terms of the expansions, in powers of η, of the eigenvalues of ρ_{AB}^Γ.

A. Laser intensity threshold for entanglement

We show here that, as a consequence of the above obtained results, A and B are entangled for low enough laser intensities. Using expression (9), the eigenvalues of ρ_{AB}^Γ can be evaluated up to second order in η. One of them is close to 1, and the others, denoted λ_q in the following, are small, since Eq. (9) with $\eta = 0$, gives $\rho_{\text{AB}}^{(0)} = |\mathcal{G}\rangle_{\text{AB}\,\text{AB}}\langle\mathcal{G}|$. Writing $\rho_{\text{AB}}^\Gamma|\varphi_q\rangle = \lambda_q|\varphi_q\rangle$, and expanding ρ_{AB}^Γ, $|\varphi_q\rangle$ and λ_q, in powers of η, with $\lambda_q^{(0)} = 0$, lead to $\lambda_q^{(1)} = 0$, and $(V + V^\dagger)|\varphi_q\rangle^{(0)} = \lambda_q^{(2)}|\varphi_q\rangle^{(0)}$, where the operator V is given by

$$V = \sum_{\mu\leqslant n_{\text{A}}<\nu\leqslant n_{\text{A}}+n_{\text{B}}} v_{\mu\nu}|\mu\rangle_{\text{AB}\,\text{AB}}\langle\nu|, \tag{13}$$

with n_{A} and n_{B} the numbers of atoms, suitably numbered, of systems A and B, respectively, and $|\mu\rangle_{\text{AB}} = \sigma_\mu^\dagger\otimes_{\nu\leqslant n_{\text{A}}+n_{\text{B}}}|g\rangle_\nu$. The complete expression of the second-order matrix $(\rho_{\text{AB}}^\Gamma)^{[2]}$ can be found in Appendix B.

The eigenvalues of the Hermitian operator $V + V^\dagger$ are real. Since it is traceless, some of them are negative as soon as $V \neq 0$. More precisely, the nonzero eigenvalues of $V + V^\dagger$ are $\pm\Lambda_q^{1/2}$ where Λ_q are the nonzero eigenvalues of both positive operators $VV^\dagger$ and $V^\dagger V$. Consequently, A and B are either uncorrelated or entangled. This is similar to the pure state case, and results from the fact that, up to second order, ρ_{AB} coincides with a pure state, as discussed above. In other words, any two subgroups of atoms are generically entangled for small enough

η. The opposite limit, $\Omega \gg \Gamma$, corresponds to the saturation regime, where $\rho_{\rm AB}$ is proportional to the identity matrix, and hence A and B are uncorrelated. Thus, there is a laser intensity threshold, that depends on A and B, where $\rho_{\rm AB}$ goes from entangled to separable.

B. Dilute regime

In the general case, determining the value of η above which $\rho_{\rm AB}$ becomes separable, requires solving Eq. (7) for finite η, which is not straightforward, even for only two atoms [8]. Moreover, for more than two atoms, there is no simple necessary and sufficient condition for entanglement [31]. However, for atoms separated by distances much larger than k_0^{-1}, which is of the order of 0.1 μm for ω_0 of some eV, a laser intensity below which A and B are certainly entangled, can be evaluated. In this dilute regime, Eqs. (11) can be solved perturbatively in the coefficients $z_{\mu\nu} \sim k_0^{-1}|\mathbf{r}_\mu - \mathbf{r}_\nu|^{-1}$ with $\mu \neq \nu$. This leads to the dominant contribution

$$v_{\mu\nu} = -4 z_{\mu\nu}(1-2i\delta)^{-3}\big(w_\mu^2 + w_\nu^2\big), \tag{14}$$

to the matrix elements of operator (13). Note that the correlations between atoms μ and ν are then the same in the presence and absence of the other atoms. We also remark that atoms not illuminated by the laser beam are also entangled for sufficiently low laser intensities. For such atoms, in the dilute regime, $v_{\mu\nu} \propto \sum_\xi z_{\mu\xi} z_{\nu\xi} w_\xi^2$, where the sum runs over the atoms in the laser field.

The eigenvalue λ_q expands, in powers of η, as $\lambda_q = \eta^2 \lambda_q^{(2)} + \eta^4 \lambda_q^{(4)} + \cdots$, where $\lambda_q^{(2)}$ is an eigenvalue of $V + V^\dagger$. Since λ_q is positive for large η, it changes sign for a certain value Ω_q of Ω, for negative $\lambda_q^{(2)}$. As the matrix elements of V are given by Eq. (14), $\lambda_q^{(2)}$ is small in the dilute regime considered here. On the contrary, $\lambda_q^{(4)}$ attains a finite value in the limit of vanishing $z_{\mu\nu}$. We find the positive asymptotic value

$$\lambda_q^{(4)} = (1/4+\delta^2)^{-2} \sum_{\mu \leqslant n_{\rm AB}} |w_\mu|^4 |{}_{\rm AB}\langle \mu|\varphi_q\rangle^{(0)}|^2, \tag{15}$$

where $n_{\rm AB} = n_{\rm A} + n_{\rm B}$, and $|\varphi_q\rangle^{(0)}$ is the eigenstate of $V + V^\dagger$ corresponding to $\lambda_q^{(2)}$; see Appendix C. This leads, for negative $\lambda_q^{(2)}$, to $\Omega_q \simeq \Gamma[|\lambda_q^{(2)}|/\lambda_q^{(4)}]^{1/2}$. As long as $\Omega < \max_q \Omega_q$, at least one eigenvalue λ_q is negative, and hence A and B are necessarily entangled.

C. Long-range entanglement

To study more quantitatively long-range entanglement, we consider two regions of characteristic size L, separated by a large distance $D \gg k_0 L^2$, and assume that, in these areas, the laser beam is essentially a plane wave of wave vector $\mathbf{K}$. Systems A and B consist of the atoms lying in these regions; see Fig. 1. In this case, Eqs. (5), (13), and (14) give

$$V = \frac{3i \sin^2\theta\, e^{ik_0 D}}{k_0 D (1-2i\delta)^3} \sum_{\mu \leqslant n_{\rm A} < \nu \leqslant n_{\rm AB}} |\tilde{\mu}\rangle\langle\tilde{\nu}|\big(w_\mu^2 + w_\nu^2\big), \tag{16}$$

where $w_\mu = \exp(i\mathbf{K}\cdot\mathbf{r}_\mu - i\omega t)$, θ is the angle between $\mathbf{d}$ and the approximate line joining A and B, and $|\tilde{\mu}\rangle = \exp(-ik_0 \mathbf{e}\cdot\mathbf{r}_\mu)|\mu\rangle_{AB}$ with $\mathbf{e}$ the unit vector pointing from A to B. Noting that this operator can be written in terms of four kets, one finds two negative eigenvalues $\lambda_q^{(2)}$. For randomly distributed atoms and large enough numbers $n_{\rm A}$ and $n_{\rm B}$, these two negative $\lambda_q^{(2)}$ are practically equal; see Appendix D. Since $|w_\mu| = 1$ for all the atoms of A and B, the sum in expression (15) reduces to 1. Finally, using the evaluation of Ω_q discussed at the end of the previous paragraph, one finds that A and B are necessarily entangled for

$$\Omega < \frac{\sqrt{3}}{2}\Gamma(1+4\delta^2)^{1/4}\left(\frac{D_0}{D}\right)^{1/2}, \tag{17}$$

where $D_0 = k_0^{-1}(n_{\rm A} n_{\rm B})^{1/2}\sin^2\theta$. The negativity $\mathcal{N}$ vanishes for Ω equal to the right side of this inequality, and also as η goes to zero. In the dilute regime considered here, it reaches a maximum for Ω equal to the right side of Eq. (17) divided by $\sqrt{2}$, which is

$$\begin{aligned}\mathcal{N}_{\rm max} &= \frac{9}{32}(1+4\delta^2)^{-1}\left(\frac{D_0}{D}\right)^2 \\ &= 32(1+4\delta^2)^{-2}\eta_{\rm max}^4,\end{aligned} \tag{18}$$

where $\eta_{\rm max}$ is the value of the ratio $\eta = \Omega/2\Gamma$ at the maximum. The attainable values of negativity are thus essentially limited by the validity of the low-laser-intensity perturbative approach we use.

As the distance between systems A and B increases, the interval of laser amplitudes that lead to nonzero negativity shrinks, and the maximum negativity $\mathcal{N}_{\rm max}$ diminishes. However, since D appears in Eqs. (17) and (18), divided by D_0, the unfavorable impact of increasing the distance can be counterbalanced by enlarging the numbers $n_{\rm A}$ and $n_{\rm B}$. An interesting consequence is that, all the other parameters, including D and the laser amplitude Ω, being fixed, big enough groups of atoms are necessarily entangled. Let us examine this point more carefully. Assuming that the atoms are uniformly distributed, and that A and B are cubes of edge length L, $n_{\rm A/B} = (L/d)^3$ where d is the mean interparticle distance, and Eq. (17) can be rewritten as

$$L > d(1+4\delta^2)^{-1/6}(k_0 D)^{1/3}(2\Omega/\sqrt{3}\Gamma\sin\theta)^{2/3}. \tag{19}$$

For L satisfying this inequality, A and B are certainly entangled, whereas the negativity vanishes for smaller groups of atoms. Note that the above results have been obtained under the condition $D \gg k_0 L^2$, which can be fulfilled together with Eq. (19) only if D is large enough. With $k_0^{-1} \simeq 0.1$ μm, $d \simeq 1$ μm, $D \simeq 1$ μm (1 cm), $\theta \simeq \pi/2$, $\delta \simeq 0$, and $\Omega/\Gamma \simeq 0.1$, Eq. (19) gives a lower bound of about 50 μm (10 μm). The corresponding number $n_{\rm A/B}$ of atoms is of the order of 10^5 (10^3). We finally discuss the influence of the laser detuning. As δ is increased, the bound given by Eq. (17) grows, and that given by Eq. (19) decreases. However, our resonant approach, based on Eq. (7), is valid only for not too large δ. Moreover, the reachable values of negativity vanish with increasing δ; see Eq. (18).

V. CONCLUSION

In summary, we have shown that two groups of two-level atoms, A and B, can be entangled by a weak resonant laser field, even if the distance between them is macroscopic, and even in the presence of surrounding identical atoms. In a dilute regime, for far separated A and B in a plane-wave laser beam, we have determined a value of the laser amplitude below which A and B are certainly entangled, and the maximum negativity that can be reached by varying the laser amplitude. They both diminish with increasing distance between A and B. But these tendencies can be counterbalanced by enlarging the sizes of A and B. Consequently, for given laser intensity and distance between the two groups of atoms, they are necessarily entangled if their size exceeds a certain value.

In this work, we assumed that the motion of the atoms is slow enough that its impact on the dynamics of their internal state can be disregarded, which, depending on the atomic mass, can be valid for temperatures of the order of 10 K. A natural extension of our study would be to examine how the found laser-induced entanglement depends on the atoms' velocities for higher temperatures, and whether it disappears at some temperature. The quantitative results presented for very distant groups of atoms have been derived in the dilute regime. It would be of interest to determine how general they are, especially the positive impact of enlarging the number of considered atoms. We finally remark that, though we focus on atoms in this paper, the studied entanglement mechanism may be relevant to other physical realizations of qubits, such as nuclear spins, coupled to a common environment, and to oscillating fields.

APPENDIX A: PERTURBATIVE SOLUTION OF EQ. (7)

With $\rho_p^{(0)} = \delta_{p0}|\mathcal{G}\rangle\langle\mathcal{G}|$, the recursive relation (8) gives

$$\rho_p^{(1)} = \delta_{p1}\sum_\mu \tilde{u}_\mu|\mu\rangle\langle\mathcal{G}| + \delta_{p\,-1}\sum_\mu \tilde{u}_\mu^*|\mathcal{G}\rangle\langle\mu|, \qquad \text{(A1)}$$

where the components $\tilde{u}_\mu$ obey Eq. (11) with $t = 0$. In deriving this result, we used the fact that the matrix elements $|\langle\mu\nu|\rho_{\pm1}|\xi\rangle| < (\langle\mu\nu|\rho_0|\mu\nu\rangle\langle\xi|\rho_0|\xi\rangle)^{1/2}$ are at least of second order. They are actually of third order.

Using again Eq. (8), we find

$$\rho_p^{(2)} = \delta_{p0}\left(-\sum_\mu |u_\mu|^2|\mathcal{G}\rangle\langle\mathcal{G}| + \sum_{\mu,\nu}\tilde{u}_\mu\tilde{u}_\nu^*|\mu\rangle\langle\nu|\right)$$
$$+\delta_{p2}\sum_{\mu<\nu}s_{\mu\nu}|\mu\nu\rangle\langle\mathcal{G}| + \delta_{p\,-2}\sum_{\mu<\nu}s_{\mu\nu}^*|\mathcal{G}\rangle\langle\mu\nu|, \qquad \text{(A2)}$$

where the components $s_{\mu\nu}$ obey $\sum_\xi(z_{\mu\xi}\tilde{s}_{\xi\nu} + z_{\nu\xi}\tilde{s}_{\xi\mu}) - 2i\delta s_{\mu\nu} = i\tilde{u}_\mu w_\nu + i\tilde{u}_\nu w_\mu$ with $\tilde{s}_{\mu\nu}$ equal to 0 for $\mu = \nu$, $s_{\mu\nu}$ for $\mu < \nu$, and $s_{\nu\mu}$ for $\mu > \nu$. With the help of the first equality of Eqs. (11), it can be shown that $v_{\mu\nu} = (s_{\mu\nu} - \tilde{u}_\mu\tilde{u}_\nu)\exp(-2i\omega t)$ satisfies the second equality of Eqs. (11). Finally, the atoms state $\rho = \sum_p \rho_p\exp(-ip\omega t)$, can be written, up to second order, under the form (9).

APPENDIX B: EXPRESSION OF $(\rho_{\mathrm{AB}}^\Gamma)^{[2]}$

The Fourier components of the second-order state $\rho_{\mathrm{AB}}^{[2]}$ of two atomic subensembles A and B are readily obtained from Eqs. (A1) and (A2) by performing a partial trace. They are given by Eqs. (A1) and (A2) with sums running only over the atoms $\mu \leqslant n_{\mathrm{AB}} = n_{\mathrm{A}} + n_{\mathrm{B}}$. Consequently, the only nonvanishing matrix elements of its partial transpose are

$$\langle\mathcal{G}|(\rho_{\mathrm{AB}}^\Gamma)^{[2]}|\mathcal{G}\rangle = 1 - \eta^2\sum_{\mu\leqslant n_{\mathrm{AB}}}|u_\mu|^2, \qquad \text{(B1)}$$

$$\begin{aligned}\langle\mathcal{G}|(\rho_{\mathrm{AB}}^\Gamma)^{[2]}|\mu\rangle &= \eta u_\mu^* \quad \text{for} \quad \mu \leqslant n_{\mathrm{A}}\\ &= \eta u_\mu \quad \text{for} \quad n_{\mathrm{A}} < \mu,\end{aligned} \qquad \text{(B2)}$$

$$\begin{aligned}\langle\mu|(\rho_{\mathrm{AB}}^\Gamma)^{[2]}|\nu\rangle &= \eta^2 u_\mu u_\nu^* \quad \text{for} \quad \mu,\nu \leqslant n_{\mathrm{A}}\\ &= \eta^2 u_\mu^* u_\nu \quad \text{for} \quad n_{\mathrm{A}} < \mu,\nu\\ &= \eta^2(u_\mu u_\nu + v_{\mu\nu}) \quad \text{for} \quad \mu \leqslant n_{\mathrm{A}} < \nu\\ &= \eta^2(u_\mu^* u_\nu^* + v_{\nu\mu}^*) \quad \text{for} \quad \nu \leqslant n_{\mathrm{A}} < \mu,\end{aligned} \qquad \text{(B3)}$$

$$\begin{aligned}\langle\mathcal{G}|(\rho_{\mathrm{AB}}^\Gamma)^{[2]}|\mu\nu\rangle &= \eta^2(u_\mu^* u_\nu^* + v_{\mu\nu}^*) \quad \text{for} \quad \mu < \nu \leqslant n_{\mathrm{A}}\\ &= \eta^2(u_\mu u_\nu + v_{\mu\nu}) \quad \text{for} \quad n_{\mathrm{A}} < \mu < \nu\\ &= \eta^2 u_\mu^* u_\nu \quad \text{for} \quad \mu \leqslant n_{\mathrm{A}} < \nu,\end{aligned} \qquad \text{(B4)}$$

$\langle\mu|(\rho_{\mathrm{AB}}^\Gamma)^{[2]}|\mathcal{G}\rangle = \langle\mathcal{G}|(\rho_{\mathrm{AB}}^\Gamma)^{[2]}|\mu\rangle^*$, and $\langle\mu\nu|(\rho_{\mathrm{AB}}^\Gamma)^{[2]}|\mathcal{G}\rangle = \langle\mathcal{G}|(\rho_{\mathrm{AB}}^\Gamma)^{[2]}|\mu\nu\rangle^*$, where $|\mathcal{G}\rangle$, $|\mu\rangle$, and $|\mu\nu\rangle$ must be understood here as $|\mathcal{G}\rangle = \otimes_{\xi\leqslant n_{\mathrm{AB}}}|g\rangle_\xi$, $|\mu\rangle = \sigma_\mu^\dagger|\mathcal{G}\rangle$, and $|\mu\nu\rangle = \sigma_\mu^\dagger\sigma_\nu^\dagger|\mathcal{G}\rangle$, with $\mu < \nu \leqslant n_{\mathrm{AB}}$.

APPENDIX C: EVALUATION OF $\lambda_q^{(4)}$ IN THE DILUTE REGIME

Writing $\rho_{\mathrm{AB}}^\Gamma|\varphi_q\rangle = \lambda_q|\varphi_q\rangle$, and expanding $\rho_{\mathrm{AB}}^\Gamma$, $|\varphi_q\rangle$, and λ_q, in powers of η, with $\lambda_q^{(0)} = 0$, give $\lambda_q^{(1)} = \lambda_q^{(3)} = 0$, $(V + V^\dagger)|\varphi_q\rangle^{(0)} = \lambda_q^{(2)}|\varphi_q\rangle^{(0)}$, where the operator V is given by Eq. (13), and, after a lengthy but straightforward derivation,

$$\lambda_q^{(4)} = \lambda_q^{(2)}\sum_{\mu<\nu}\left|\tau_q^{\mu\nu}\right|^2 - \left|\tau_q^{\mathcal{G}}\right|^2\left(\lambda_q^{(2)} + \sum_\mu |u_\mu|^2\right)$$
$$+ \langle\phi_q|(\rho_{\mathrm{AB}}^\Gamma)^{(4)}|\phi_q\rangle + 2\mathrm{Re}\left[\tau_q^{\mathcal{G}}\langle\phi_q|(\rho_{\mathrm{AB}}^\Gamma)^{(3)}|\mathcal{G}\rangle\right], \qquad \text{(C1)}$$

where $|\phi_q\rangle = |\varphi_q\rangle^{(0)}$, $\tau_q^k = \langle k|\varphi_q\rangle^{(1)}$, and the sums run only over the atoms $\mu \leqslant n_{\mathrm{AB}}$. In this appendix, as in the previous one, $|\mathcal{G}\rangle$, $|\mu\rangle$, and $|\mu\nu\rangle$ must be understood as $|\mathcal{G}\rangle = \otimes_{\xi\leqslant n_{\mathrm{AB}}}|g\rangle_\xi, |\mu\rangle = \sigma_\mu^\dagger|\mathcal{G}\rangle$, and $|\mu\nu\rangle = \sigma_\mu^\dagger\sigma_\nu^\dagger|\mathcal{G}\rangle$, with $\mu < \nu \leqslant n_{\mathrm{AB}}$. The only component of $|\varphi_q\rangle^{(1)}$ required for our purpose is $\tau_q^{\mathcal{G}} = -\sum_\mu \hat{u}_\mu\langle\mu|\phi_q\rangle$, where $\hat{u}_\mu = u_\mu^*$ for $\mu \leqslant n_{\mathrm{A}}$, and u_μ for $\mu > n_{\mathrm{A}}$.

We are concerned with the value of $\lambda_q^{(4)}$ in the limit of infinitely distant atoms, in which ρ_{AB} converges to the uncorrelated state $\rho_{\mathrm{AB}}^{(\mathrm{dl})} = \otimes_{\mu\leqslant n_{\mathrm{AB}}}\rho_\mu^{(\mathrm{dl})}$ where

$$\rho_\mu^{(\mathrm{dl})} = (1 - p_\mu)\sigma_\mu\sigma_\mu^\dagger + p_\mu\sigma_\mu^\dagger\sigma_\mu + c_\mu\sigma_\mu^\dagger + c_\mu^*\sigma_\mu, \qquad \text{(C2)}$$

with $p_\mu = \eta^2|w_\mu|^2/(1/4 + \delta^2 + 2\eta^2|w_\mu|^2)$, and $c_\mu = (i/2 - \delta)p_\mu/\eta w_\mu^*$. Expanding this density matrix in η gives $(\rho_{\mathrm{AB}}^\Gamma)^{(3)}$ and $(\rho_{\mathrm{AB}}^\Gamma)^{(4)}$ in the infinitely dilute regime. Using the resulting expressions, the vanishing of $\lambda_q^{(2)}$ in this asymptotic regime, and equality (C1), leads to Eq. (15).

APPENDIX D: DIAGONALIZATION OF THE OPERATOR $V + V^\dagger$ FOR V GIVEN BY EQ. (16)

The operator (16) can be written in the form

$$V = |\phi_+\rangle\langle\phi'_-| + |\phi'_+\rangle\langle\phi_-|. \tag{D1}$$

The components of the above kets are $\langle\tilde{\mu}|\phi_\pm\rangle = (1 \pm \zeta_\mu)/2$, and $\langle\tilde{\mu}|\phi'_\pm\rangle = x_\mu^\pm(1 \pm \zeta_\mu)/2$, where $\zeta_\mu = 1$ for $\mu \leqslant n_A$, and -1 for $\mu > n_A$, $x_\mu^+ = x\exp(ik_0D + 2i\mathbf{K}\cdot\mathbf{r}_\mu - 2i\omega t)$ with $x = 3i\sin^2\theta/k_0D(1-2i\delta)^3$, and $x_\mu^- = (x_\mu^+)^*$. To find the nonzero λ obeying the eigenvalue equation $(V + V^\dagger)|\varphi\rangle = \lambda|\varphi\rangle$, we expand $|\varphi\rangle$ on the basis $\{|\phi_+\rangle, |\phi'_+\rangle, |\phi_-\rangle, |\phi'_-\rangle\}$. This leads to the characteristic equation

$$\lambda^4 - 2y[1 + \mathrm{Re}(s_A s_B^*)]\lambda^2 + y^2[1 - |s_A|^2][1 - |s_B|^2] = 0, \tag{D2}$$

where $y = n_A n_B |x|^2$, $s_A = n_A^{-1}\sum_{\mu\leqslant n_A}\exp(2i\mathbf{K}\cdot\mathbf{r}_\mu)$, and $s_B = n_B^{-1}\sum_{\mu>n_A}\exp(2i\mathbf{K}\cdot\mathbf{r}_\mu)$.

For randomly distributed atoms and large enough numbers n_A and n_B, s_A and s_B are negligible, and the above equation simplifies to $(\lambda^2 - y)^2 = 0$. This last result can be derived more directly as follows. The relations $s_A, s_B \ll 1$ can be rewritten as $\langle\phi_\pm|\phi'_\pm\rangle \simeq 0$. When these products vanish, it is immediate to see that the nonzero eigenvalues of both $VV^\dagger$ and $V^\dagger V$, which are the squares of the nonzero eigenvalues of $V + V^\dagger$, are $\langle\phi_\pm|\phi_\pm\rangle\langle\phi'_\mp|\phi'_\mp\rangle = y$.

[1] T. Yu and J. H. Eberly, Phys. Rev. Lett. **93**, 140404 (2004).
[2] D. Braun, Phys. Rev. Lett. **89**, 277901 (2002).
[3] L. Jakóbczyk, J. Phys. A: Math. Gen. **35**, 6383 (2002).
[4] F. Benatti, R. Floreanini, and M. Piani, Phys. Rev. Lett. **91**, 070402 (2003).
[5] Z. Ficek and R. Tanaś, Phys. Rev. A **74**, 024304 (2006).
[6] D. P. S. McCutcheon, A. Nazir, S. Bose, and A. J. Fisher, Phys. Rev. A **80**, 022337 (2009).
[7] Ö. Çakir, A. A. Klyachko, and A. S. Shumovsky, Phys. Rev. A **71**, 034303 (2005).
[8] K. Almutairi, R. Tanaś, and Z. Ficek, Phys. Rev. A **84**, 013831 (2011).
[9] S. Camalet, Eur. Phys. J. B **84**, 467 (2011).
[10] B. Bellomo and M. Antezza, Europhys. Lett. **104**, 10006 (2013).
[11] B. Bellomo and M. Antezza, New J. Phys. **15**, 113052 (2013).
[12] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Processus d'Interaction entre Photons et Atomes* (CNRS Editions, Paris, 1988).
[13] R. H. Dicke, Phys. Rev. **93**, 99 (1954).
[14] R. H. Lehmberg, Phys. Rev. A **2**, 889 (1970).
[15] T. Wilk, A. Gaëtan, C. Evellin, J. Wolters, Y. Miroshnychenko, P. Grangier, and A. Browaeys, Phys. Rev. Lett. **104**, 010502 (2010).
[16] X. L. Zhang, L. Isenhower, A. T. Gill, T. G. Walker, and M. Saffman, Phys. Rev. A **82**, 030306(R) (2010).
[17] A. S. Parkins, E. Solano, and J. I. Cirac, Phys. Rev. Lett. **96**, 053602 (2006).
[18] C. A. Muschik, E. S. Polzik, and J. I. Cirac, Phys. Rev. A **83**, 052312 (2011).
[19] F. Reiter, M. J. Kastoryano, and A. S. Sørensen, New J. Phys. **14**, 053022 (2012).
[20] H. Krauter, C. A. Muschik, K. Jensen, W. Wasilewski, J. M. Petersen, J. I. Cirac, and E. S. Polzik, Phys. Rev. Lett. **107**, 080503 (2011).
[21] C. A. Muschik, H. Krauter, K. Jensen, J. M. Petersen, J. I. Cirac and E. S. Polzik, J. Phys. B: At. Mol. Opt. Phys. **45**, 124021 (2012).
[22] M. Macovei, J. Evers, and C. H. Keitel, J. Mod. Opt. **57**, 1287 (2010).
[23] S. Camalet, Eur. Phys. J. B **86**, 176 (2013).
[24] R. H. Lehmberg, Phys. Rev. A **2**, 883 (1970).
[25] J. Dalibard and C. Cohen-Tannoudji, J. Opt. Soc. Am. B **2**, 1707 (1985).
[26] M. J. Stephen, J. Chem. Phys. **40**, 669 (1964).
[27] M. Trippenbach, B. Gao, J. Cooper, and K. Burnett, Phys. Rev. A **45**, 6539 (1992).
[28] C. C. Gerry, Phys. Rev. A **53**, 4583 (1996).
[29] S. J. van Enk, Phys. Rev. A **72**, 064306 (2005).
[30] K. S. Choi, H. Deng, J. Laurat, and H. J. Kimble, Nature (London) **452**, 67 (2008).
[31] R. Horodecki, P. Horodecki, M. Horodecki, and K. Horodecki, Rev. Mod. Phys. **81**, 865 (2009).