Vacuum fluctuations induced entanglement between two mesoscopic systems

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**Abstract.** We study the dynamics of a pair of molecular ensembles trapped inside a superconducting resonator through which they are strongly coupled via a microwave field mode. We find that entanglement can be generated via ‘vacuum fluctuations’ even when the molecules and cavity field are initially prepared in their ground state. This entanglement is created in a relatively short time and without the need for further manipulation of the system. It therefore provides a convenient scheme to entangle two mesoscopic systems, and may well be useful for quantum information processing.
Vacuum fluctuations can have important physical consequences, for example, in the Casimir effect [1] and Hawking radiation [2]. In particular, van der Waals interactions, i.e. attractive long-ranged interaction between neutral atoms or molecules, are also a version of the Casimir effect. It is an interesting question as to how vacuum fluctuations might be used to influence the properties of quantum entanglement between two systems. Quantum entanglement is a fundamental concept in quantum mechanics [3] and can represent the physical resource for use in quantum information processing [4]. In fact, it has recently been shown how one may generate entanglement between a pair of particles via the vacuum modes of the radiation field [5]–[7].

In this paper, we study how vacuum fluctuations may be used to induce quantum entanglement between two mesoscopic systems, i.e. polar molecular ensembles [8] that are placed in a cavity, and hence strongly coupled by a single microwave mode. Recently, Rabl et al [9] have proposed the realization of a quantum memory using such ensembles of polar molecules inside a superconducting resonator [10]. Polar molecules are favoured as the energy difference between two internal states of a polar molecule is of the order of GHz, and they have significant electric dipole moments. A strong coupling to a microwave field via a transmission line can thus be achieved. In addition, low-lying collective excitations can be coupled to the field, and the enhanced coupling to them, which scales as $\sqrt{N}$, where $N$ is the number of molecules in the ensemble, can be exploited.

The dynamics of vacuum fluctuations is hard to observe in ordinary systems. To show why this is so, we start the simple case of a two-level atom interacting with a quantized field. Conventionally, we use the Jaynes–Cummings model [11] in the interaction picture ($\hbar = 1$), Hamiltonian $H = g' \sigma_- e^{i(\omega' - \omega_0)t} + \sigma_+ e^{-i(\omega' - \omega_0)t}$, to describe a two-level system $\sigma_\pm$ coupled to a quantized field $b$, for $\omega'_0$, $\omega'$ and $g'$ are an energy difference between two-level atom, the frequency of the field and the Rabi frequency, respectively. The rotating wave approximation (RWA) can usually be used because the two counter-rotating terms, $b\sigma_-$ and $\sigma_+ b^\dagger$, can be neglected; they carry a fast oscillation with the high frequency $\omega' + \omega_0$. The RWA is, therefore, an excellent approximation for the optical frequency regime in the weak Rabi coupling limit. Clearly, this Hamiltonian will produce no evolution in the atoms and the photon field if they both start in the respective ground states. However, this approximation breaks down if the Rabi frequency $g'$ is comparable with the frequencies $\omega'$ and $\omega'_0$. In fact, the RWA is completely inadequate to describe the physical situation of a large number of molecules interacting with a microwave field in the strong coupling regime. It is thus necessary to go beyond RWA and, in essence, study the role of vacuum modes on the dynamics of the coupled molecule–field system.

We consider the case where molecules and the photon field are initially prepared in their ground states and show how the counter-rotating terms in the Hamiltonian do indeed induce quantum correlations between the molecules. The two ensembles of molecules exchange energy with the vacuum field due to the counter-rotating terms. In physical terms we would say that this proceeds via virtual excitations of the cavity mode. We should bear in mind that these vacuum mode processes can be enhanced if collective excitations of the ensembles are used. As the dynamics takes place, the two molecular ensembles becomes entangled as the molecules are effectively coupled through the exchange of energy via the vacuum mode of the cavity. We will show that this entanglement can be generated in a comparatively short time. This result provides a novel route to entangle two mesoscopic systems. Relaxation and decoherence effects should

The effect of vacuum fluctuations is of course observed in static phenomena such as the Lamb shift.
Two molecular ensembles, separated by a distance $d$, are trapped inside a superconducting resonator and interact with a single-mode microwave field along the stripline.

also be modest as the system is prepared in a vacuum state; a potentially crucial advantage for practical applications.

In this paper, we suppose the molecular ensembles are placed at a distance $d$ apart in an effectively one-dimensional resonator as shown in figure 1. For example, the two states of the diatomic molecules such as CaF or CaBr, produced by the splitting of a rotational ground level by hyperfine interactions are chosen as the internal molecular states [9]. They have an energy difference $\omega$, and are coupled to a single-mode microwave field with a frequency $\omega_0$. Each of the molecules can be described by a spin-half particle $\vec{\sigma}_i$, and hence a collection of such spin-half particles can be described by a collective angular momentum operator $\vec{J} = \sum_{i=1}^{N} \vec{\sigma}_i$, when $N$ is the number of molecules in one of the ensembles. The wavelength of microwave radiation will be much longer than the size of molecular ensemble. We can therefore assume the microwave field coupling to all molecules is the same. We can now write down the Hamiltonian $H$: 

$$ H = \sum_{i=1}^{2} \omega_0 a_\dagger a + \omega J_z + g_i (a + a_\dagger) (J_+ + J_-), $$ (1)

Here, $a_\dagger$ and $a$ are the creation and annihilator operators of the cavity mode, $J_z$ and $J_x$ are the angular momentum operators to describe the collective inversion and transition for the $i$th ensemble, respectively, and $i = 1, 2$. The molecule–photon interaction strength is denoted by $g_i$ for the $i$th ensemble and they differ with a relative phase $\phi = 2\pi \omega_0 d/c$ between the field and two ensembles, where $c$ is the speed of the microwave photon. For simplicity, the magnitude of the two Rabi coupling strengths is chosen to be the same, $|g_1| = |g_2|$, and $\phi \approx 0$. We consider the case where the molecules and photon field are in resonance, i.e. $\omega = \omega_0$, which is the optimal condition to observe the effect of the vacuum coupling. We note that the Hamiltonian $H$ has the same form as the Hamiltonian of the Dicke model without the RWA [12]. The analysis we present here applies to thermal atomic ensembles as well as condensates. Condensates would have the advantage of longer coherence times but also introduce nonlinear dynamical problems.

To study the quantum dynamics of this system, we need to find the eigenstates of the whole system. The Hilbert space of this system is extremely large for even a modest number of molecules.

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5 Here the static dipolar interactions between the molecules are neglected in the low density of molecular gases because they are very small compared with the strength of the molecule–field interaction [9].

6 Generation of nontrivial quantum states in atomic condensates due to nonlinear interactions has been discussed in [13]. However, there are no significant changes of scattering properties due to the induced dipolar interaction at the temperature ($T \sim 1$ mK) [9].
of molecules. We can, however, consider that the time evolution involves only the low-lying excitations of the molecules. We can make this approximation based on the Holstein–Primakoff transformation (HPT) [14] which enables us to express angular momentum operators in terms of harmonic oscillators. In this manner, we can take the leading approximation and map an angular momentum operator into a harmonic oscillators by taking the lowest order version of the HPT [15, 16]. We then get $J_i \approx \sqrt{\omega N_i} \frac{x_i}{\sqrt{2}}, J_i \approx -\sqrt{\omega} \frac{p_i}{\sqrt{2}}, J_i \approx \frac{1}{2} (p_i^2 + \omega^2 x_i^2) / 2\omega - N_i / 2$ where $x_i$ and $p_i$ are the position and momentum operators, and $N_i$ is the number of molecules in the cavity. This approximation is valid as long as $(p_i^2 + \omega^2 x_i^2) \ll \omega N_i$ [15, 16]. It should be a very good approximation if the number of molecules is sufficiently large [9].

For convenience, we write the cavity field operators in the phase-space representation: $x_c = (a^\dagger + a) / \sqrt{2\omega_0}$ and $p_c = i \sqrt{\omega_0}(a^\dagger - a) / \sqrt{2}$. The Hamiltonian of system can then be rewritten in the form:

$$H' = \frac{1}{2} \sum_{i=1}^{2} (p_i^2 + p_{i1}^2 + \omega_{i}^2 x_i^2 + \omega_{i}^2 x_{i1}^2 + 4g_i \sqrt{N_i} \omega_0 x_i x_{i1}).$$

We now want to determine the dynamics as the ensembles interact and become entangled. This problem is clearly related to what finding the entanglement between two harmonic oscillators in an open-ended harmonic chain [17].

This harmonic system will be in a Gaussian state, which allows us to quantify the general bipartite entanglement of the system. The density matrix of a Gaussian state can be completely determined by the second-order moments of the position and momentum operators of the system. We just need to study the reduced density matrix of the molecular part to find out the entanglement produced between the two ensembles. This reduced density matrix can be determined by the second-order moments of the position and momentum operators of the system. We just need to study the reduced density matrix of the molecular part to find out the entanglement produced between the two ensembles. This reduced density matrix can be obtained by tracing out the cavity mode. The matrix elements of reduced density matrix $\rho_{a1,2}$ are $\langle X_i X_j + X_j X_i \rangle - 2\langle X_i \rangle \langle X_j \rangle$, where $X_i = x_{ai}$ or $p_{ai}$. A quantitative measure of entanglement can be obtained by using the logarithmic negativity [18] which gives us an upper bound for distinguishable entanglement. The logarithmic negativity in a Gaussian state can be found as [18, 19]

$$\ln N = -\sum_j \log_2[\text{min}(1, |\gamma_j|)],$$

where $\gamma_j$ are the symplectic eigenvalues of the matrix $\rho_{a1,2}$.

We are now ready to investigate the entanglement dynamics of this system. We consider the initial state as the ground state of molecules and cavity, i.e. the state of the decoupled harmonic oscillators. In figure 2, we plot the time evolution of the entanglement between the ensembles. The system begins with a separable state and then the entanglement grows rapidly. In fact, the quantum state of two ensembles oscillates between being separable and entangled. This is very similar to the entanglement propagation in a harmonic chain in which the two oscillators are periodically entangled and unentangled [17].

Moreover, the system achieves the first maximal entanglement within the time interval $t^* = 5^{-3} g^{-1}$ as shown in figure 2. We can estimate this time $t^*$ with the realistic parameters. If we take $g$ as $1$ MHz [9], nearly maximal entanglement can be yielded within $5$ ns. This means that a significant entanglement can be obtained rather quickly. Moreover, no further adjustment of the experimental parameters or making conditional measurements are required [20]. The timescale of this entanglement generation ($\sim 10$ ns) is much shorter than the other decoherence sources [9] such as inelastic collisions ($\kappa \sim 10 \mu$s) and photon losses ($\gamma \sim 1 \mu$s) such that the strong collective cooperativity $C = g^2 N / 2\kappa \gamma \sim 10^5$ can be achieved in this case. Entanglement can therefore be observed before decoherence effect set in showing in a natural and efficient way.
to generate quantum entanglement for two mesoscopic systems. In addition, we can see that a larger ratio of $g/\omega$ can produce a larger degree of entanglement in figure 2, clearly indicating that counter-rotating terms cannot be neglected in this strong coupling limit.

We should be aware that thermal noise is the main potential problem in entanglement production. It is of course impossible to prepare the perfect vacuum state of a molecular ensemble in an experiment at finite temperature. We now assume that an ensemble can be described as a thermal state with mean excitation number $\bar{n} = \left[\exp\left(\frac{\hbar \omega}{k_B T}\right) - 1\right]^{-1}$ [21], where $k_B$ is the Boltzmann constant and $T$ is the temperature. We can estimate $\bar{n}$ to be of the order of 0.1 to 1 when $\omega \sim 1 \text{ GHz}$ and $T \sim 10 \text{ mK}$ [9]. From this estimate, we can see that thermal effects cannot be neglected, and it is important to study their influence on entanglement. Time evolution of entanglement of the ensembles with the initial thermal states is shown in figure 3. The amount of entanglement produced is shown to be less in the cases of higher $\bar{n}$. Moreover, the longer onset time of entanglement is required as shown in the higher temperature cases. But the entanglement can still be observed even if $\bar{n}$ is as high as 0.2. This result shows that a substantial amount of quantum entanglement can be effectively produced using thermal ensembles but colder molecules result in a much better performance.

Having discussed the production of entanglement, we now study how to observe the quantum correlations. In this Gaussian system, the density matrix can be constructed if the uncertainties of these two ensembles can be obtained. This means that the entanglement of the two molecular ensembles can be determined just from the quantum uncertainties. In fact, non-resonant stimulated Raman scattering has been used to generate and verify the entanglement between two atomic ensembles [20, 22, 23]. In this scheme, the detuned Stokes pulses are used to ‘write’ the quantum information on the ground state of the ensembles of two-level atoms. The scattered Stokes photons are emitted and then create the excitations of each ensemble [20, 22]. Then, the two Stokes photon fields coming from each ensemble pass through a 50:50 beam splitter (BS) so that the two modes interfere and mix together.
Figure 3. The logarithmic negativity $\ln N$ is plotted against the dimensionless time $gt$ with $\omega = 300g$ and $N = 10^4$. The solid, dashed–dotted, dashed and dotted lines are represented $\bar{n} = 0$, 0.05, 0.1 and 0.2, respectively.

The conditional measurement of the resultant Stokes field can be performed and entangle the two atomic ensembles [20, 22]. Similarly, the anti-Stokes pulses can be applied to read the excitations of the atoms and then the entanglement can be verified by measuring the correlations of photon fields.

We now describe a method to determine the entanglement of the two molecular ensembles as shown in figure 4. We first take the molecules away from the electrode so that the height $z$ of the trapping molecules is increased above the stripline. The coupling between the microwave field and the molecules is switched off if the height $z$ is sufficiently large. This is because the Rabi coupling strength is inversely proportional to the distance between the trapped molecules and electrode [8]. This forms as an effective switch for molecule–photon interactions by controlling the distance between the molecules and stripline. We can thus apply two anti-Stokes pulses on these two ensembles without the cavity. The infrared radiation can be used for the pulses which have been used extensively in vibrational spectroscopy [24]. This is because the vibration–rotation spectra for the heteronuclear diatomic molecules have very simple selection rules for the rotation and vibration transitions without any change in the electronic states. The Raman scattering can be achieved by applying the detuned anti-Stokes pump field to the upper hyperfine ground state and emit anti-Stokes photons from the appropriate upper vibrational state (within the same electronic ground state)\(^7\) to the lower hyperfine ground state. Besides, we require that the Fresnel number $F = A (L \lambda_s)^{-1}$ is of the order of unity for the efficient stimulated Raman coupling [25], where $A$ and $L$ are the cross-section area and the length of the molecular ensemble, respectively, and $\lambda_s$ is the wavelength of the anti-Stokes photon. The Fresnel number $F$ is roughly equal to one if $A \sim 10^{-4}$ cm\(^2\), $L \sim 0.1$ cm and $\lambda_s \sim 10^{-3}$ cm.

To read out the excitation state of the molecules, we apply an anti-Stokes pulse to each ensemble. In the Heisenberg picture, the output beam, after passing through the ensemble [22],

\(^7\) For heteronuclear diatomic molecules, the selection rules for the vibration–rotation spectra are: the vibrational Raman transitions: $\Delta v = \pm 1$ and Raman vibration–rotation transitions: $\Delta J = 0, \pm 2$ [24].
Figure 4. (a) Schematic of homodyne detection of a single molecular ensemble. An anti-Stokes pump pulse is applied onto a molecular ensemble. The output field from the molecules is superimposed on the field from a local oscillator (LO) at a 50:50 BS and then they are detected by the detectors $D_1$ and $D_2$, respectively. (b) The output fields from two molecular ensembles are interfered with a 50:50 BS and then the fields probed using balanced homodyne detection.

is given by $a_1^O = \sqrt{\eta_i}c_i + \sqrt{1-\eta_i}a_i^I$, where $a_1^O$ and $a_i^I$ are the output field and the vacuum field operator and $\eta_i$ is the effective transmission efficiency. We can see that the output field directly carries the information of collective excitations of the molecules. Hence, the quantum state of the ensembles can be determined through the measurement of the output photon fields.

This is achieved via measurement of the field from individual ensembles as well of ‘joint’ values after mixing on a BS. We then require the detection of the individual ensembles and the two ensembles, respectively. In figure 4(a), we give a sketch of a scheme to measure the local quantum correlations of an individual ensemble by balanced homodyne detection with a LO mode with phase $\phi_l$ [11]. The moments $\langle x_i^2 \rangle$, $\langle p_i^2 \rangle$ and $\langle x_i p_i + p_i x_i \rangle$ can all be probed by appropriately adjusting the phase angle $\phi_l$. Similarly, the joint quantum correlations can be probed by this method [11]. This can be done by interfering two output fields with a 50:50 BS and then performing balanced homodyne detection as indicated in figure 4(b). The quadrature of the two modes can be thus determined.

It is noted that quantum non-demolition detection of quantum states of atomic gases can also be applied to the entanglement detection here [26]. In this scheme, the sublevels of the atoms are off-resonantly coupled to the polarization of light. In principle, the molecule–light interfaces can be formed by coupling the two ground states to the hyperfine states of the upper
vibrational states off-resonantly with the polarized light pulses. The quantum states can thus be probed through the output light [26].

In summary, we have found an efficient method to generate entanglement between two separate ensembles of molecules and proposed a method to measure it. We have also assessed the role of the finite temperature on the entanglement produced. This may well be useful for quantum information processing with molecular systems. We envisage that evaporative cooling of the trapped molecules will be realized [27] so that the temperature can be lowered and the performance of quantum memory and entanglement generation be further improved.

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