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# Photon-photon interactions in cavity electromagnetically induced transparency

M. J. Werner and A. Imamoḡlu
*Department of Electrical and Computer Engineering and Physics, University of California, Santa Barbara, California 93106-9560*

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Dissipation-free photon-photon interaction at the single-photon level is studied in the context of cavity electromagnetically induced transparency (EIT). For a single multilevel atom exhibiting EIT in the strong cavity-coupling regime, the anharmonicity of the atom-cavity system has an upper bound determined by single atom-photon coupling strength. A photon blockade effect is inferred to occur for both single- and multiatom cases from the behavior of transition rates between dressed states of the system. Numerical calculations of the second-order coherence function indicate that photon antibunching in both single- and two-atom cases are strong and comparable.

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Enhancement of dissipation-free photon-photon interactions at the few-photon level is a fundamental challenge in quantum optics. Such interactions are necessary for single-photon quantum control as in quantum logic gates. Typically, tuning close to atomic resonances simultaneously increases both (Kerr) nonlinearity and linear absorption, severely limiting the available nonlinear phase shift. In Ref. [1], a new method that is based on electromagnetically induced transparency (EIT) [2] was proposed. In this scheme linear susceptibility vanishes on resonance, making it possible to obtain as much as an eight order-of-magnitude improvement in Kerr nonlinearity as compared to conventional schemes based on three-level atoms. Recently, experimental realization of this scheme with an enhancement approaching $10^7$ using a sodium Bose-Einstein-condensate (BEC) has been reported [3]. In light of these developments, it is natural to explore the limits on achievable photon-photon interaction while maintaining EIT.

A single photon in a cavity can block the injection of a second photon due to a photon blockade effect [4,5]. The original proposal for using a cavity-EIT medium to achieve photon blockade was based on an adiabatic elimination of the atomic degrees of freedom [4]. Using a linearized analysis, Grangier *et al.* [6] have shown that the adiabatic elimination carried out in Ref. [4] is not justified due to large dispersion of the EIT medium. The correct description of photon-photon interactions in the single-photon limit requires a nonlinear analysis that treats the atom-photon interactions exactly: this is the principal goal of this Rapid Communication, where the analysis is based on the exact energy eigenstates of the coupled atom-cavity system. Under conditions of strong coupling, the anharmonicity in the spectrum of the atom-cavity molecule can lead to nonclassical photon statistics such as photon antibunching or a photon blockade effect. The latter is determined by the probability of two photons absorbed sequentially into the system, while the former is related to the conditional probability of photon emission given that a single photon was already emitted. Both effects strongly depend on the anharmonicity of the atom-cavity energy eigenstates in the doubly excited ($n=2$) manifold, or equivalently, how well can the coupled system, under EIT conditions, be described as an effective two-level system [7]. Even though our analysis focuses on determining the level shifts and linewidths in the $n=2$ manifold, we complement this analysis with a calculation of the second-order coherence function of the transmitted field.

We investigate the dependence of the photon-photon interaction on the atom number, dispersion, and the atom-cavity coupling strength. We concentrate on the situation where the cavity contains at most two photons, the atom-cavity system exhibits EIT at the cavity mode frequency, and a weak external driving field is resonant with the EIT transition so that the only state excited by the first photon (i.e., in the $n=1$ manifold) is the cavity-EIT state. The crucial advantages of this EIT scheme are the absence of one photon loss due to spontaneous emission, an arbitrarily reduced cavity decay rate for the $n=1$ manifold cavity-EIT state, and a multiatom-induced photon-photon interaction, which, under conditions of nonresonant EIT, can be comparable to a single atom. In the context of quantum logic gates, atom-atom interactions between two multilevel atoms induced via a single cavity mode have been discussed previously [8] where the role of photons and atoms is reversed compared to our scheme.

The definition of photon blockade suggests that its strength can be quantified by the transition rate $W_{1\rightarrow 2}$ from the $n=1$ manifold to the $n=2$ manifold, normalized to the $n=0 \rightarrow n=1$ transition rate. Assuming a single relatively stable state in the $n=1$ manifold, we can measure the strength of the photon blockade effect by $P=\Sigma_i W_{1\rightarrow 2i}/W_{0\rightarrow 1}$, which in turn suggests that one way to analyze the problem is to calculate transition rates between exact dressed states. An experimentally more relevant quantity is the degree of photon antibunching determined by $g^2(0)=\langle \hat{a}^{\dagger 2}\hat{a}^2\rangle/\langle \hat{a}^{\dagger}\hat{a}\rangle^2$. Even though these quantities are physically different, as described earlier, when the coupled system behaves like a single two-level system, $g^2(0)\ll 1$ implies photon blockade ($P\ll 1$).

Consider the $n=1$ manifold of the coupled atom-cavity system where the cavity mode contains at most one photon. Since we assume throughout this paper that the EIT condition is satisfied, the frequencies of the classical coupling field ($\omega_c$) and the cavity mode ($\omega_{cav}$) are chosen to satisfy two-photon resonance $\omega_{cav}-\omega_c-\omega_{21}=\omega_{31}-\delta-\omega_c-\omega_{21}=0$ (see Fig. 1). The EIT condition implies that there is no population in the upper state $|3\rangle$, which is the only atomic state with a significant decay rate relevant to the $n=1$ manifold.

 

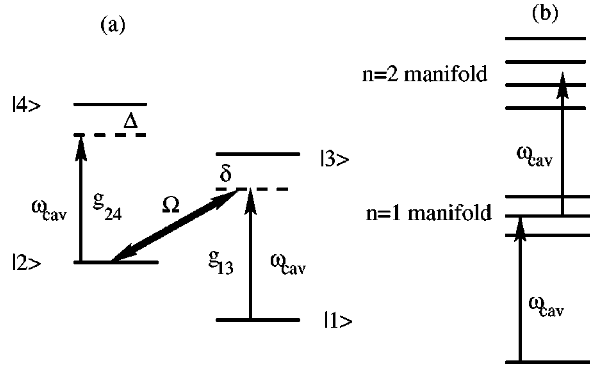


FIG. 1. (a) Internal atomic degrees of freedom of a four-level atom coupled to a single cavity mode with frequency $\omega_{cav}$ and an external laser field characterized by the Rabi frequency $2\Omega$. (b) The energy levels of the coupled atom-cavity system.

The effective interaction-picture Hamiltonian that couples $N$ four-level atoms with the single-cavity mode and the coupling field with Rabi frequency $2\Omega$ is

$$\hat{H}/\hbar = -i\kappa\hat{a}^\dagger\hat{a} + \sum_{i=1}^{N} [-i\tilde{\Gamma}_4\hat{\sigma}_{44}^i - i\tilde{\Gamma}_3\hat{\sigma}_{33}^i + \Omega(\hat{\sigma}_{32}^i + \hat{\sigma}_{23}^i) + g_{13}(\hat{a}\hat{\sigma}_{31}^i + \hat{a}^\dagger\hat{\sigma}_{13}^i) + g_{24}(\hat{a}\hat{\sigma}_{42}^i + \hat{a}^\dagger\hat{\sigma}_{24}^i)], \quad (1)$$

where $\gamma_{31}, \gamma_{32}, \gamma_4$ are atomic-level spontaneous-emission rates, $\tilde{\Gamma}_3 = (\gamma_{31} + \gamma_{32})/2 + i\delta$, $\tilde{\Gamma}_4 = \gamma_4/2 + i\Delta$, $\kappa$ is the cavity decay rate, and $\Delta = \omega_{41} - \omega_{21} - \omega_{cav}$. The externally measured photon statistics of the driven cavity are determined by the master equation $\dot{\rho} = -i[\hat{H}/\hbar, \rho] + \mathcal{E}_p[\hat{a}^\dagger - \hat{a}, \rho] + \hat{\mathcal{J}}\rho$, where $\hat{\mathcal{J}}$ is the jump superoperator for the Lindblad form and $\mathcal{E}_p$ is the strength of the weak external drive field resonant with the cavity mode. The last term of Eq. (1) can be replaced by $H_{perturb} = -\hbar\chi\hat{a}^\dagger\hat{a}\Sigma_i\hat{\sigma}_{22}^i$ in the limit $|\Delta| \gg g_{24}, \Gamma_4$, where $\chi = g_{24}^2/\Delta$.

In the case of a single atom in the cavity ($N=1$), the $n=1$ and $n=2$ manifolds (in the perturbative limit) have three eigenstates. In the limit $\tilde{\Gamma}_3 = 0$, eigenvalues $\hbar(\omega_{cav} + \epsilon_i)$ for the $n=1$ manifold are approximately given by

$$\epsilon_0^{n=1} \approx \frac{-i\kappa}{1+\alpha}, \quad \epsilon_\pm^{n=1} \approx -i\frac{\kappa}{2} \pm \Omega\sqrt{1+\alpha-(\kappa/2\Omega)^2}, \quad (2)$$

where $\alpha = g_{13}^2 N/\Omega^2$ and $\mathrm{Re}(\epsilon_0^{n=1})$ is independent of $(g_{13}, \Omega)$ and is on cavity resonance. The eigenstate corresponding to $\epsilon_0^{n=1}$,

$$|\phi_0^{n=1}\rangle = \frac{1}{\sqrt{1+\alpha}}\left[\hat{a}^\dagger - \frac{g_{13}}{\Omega}\hat{\sigma}_{21}\right]|0\rangle, \quad (3)$$

contains no component from the upper atomic state $|3\rangle$ and in the limit $\alpha \gg 1$ is predominantly atomic in character: this is the cavity-EIT trapping state. The transition rate induced by a weak probe from the ground state into $\epsilon_0^{n=1}$ is independent of $\alpha$. In the $n=2$ manifold, in the limit $g_{24}=0$, one of the eigenstates is

$$|\phi_0^{n=2}\rangle = \frac{1}{\sqrt{2+\alpha}}\hat{a}^\dagger\left[\hat{a}^\dagger - \frac{g_{13}}{\Omega}\hat{\sigma}_{21}\right]|0\rangle, \quad (4)$$

whose linewidth due to cavity decay has a weak $\alpha$ dependence of $\kappa(4+\alpha)/(2+\alpha)$. For $g_{24} \neq 0$, this energy eigenstate experiences an ac-Stark shift. Once the EIT supporting state $|\phi_0^{n=1}\rangle$ is excited, the state closest to two-photon resonance for $\delta=0$ ($|\phi_0^{n=2}\rangle$) has a perturbative level shift due to $g_{24}$ of

$$\epsilon^{n=2} \approx -\frac{g_{24}^2}{\Delta}\frac{2g_{13}^2}{2g_{13}^2+\Omega^2} = -\chi\frac{2\alpha}{1+2\alpha}, \quad (N=1) \quad (5)$$

allowing photon-blockade and photon antibunching to occur, provided $\chi > \kappa$. The level shift is largest in the limit $\alpha \gg 1$, which can be achieved by reducing $\Omega$. One should note that additional constraints on $\Omega$ exist to maintain EIT but are weak for cold alkali-metal atoms [3].

The results outlined above only consider the regime where level $|4\rangle$ can be adiabatically eliminated. In this perturbative limit, we found that the maximum value of the photon-photon interaction (as measured by the anharmonicity or level shift in analogy with an ideal nonlinear cavity) is independent of $\alpha$ and is given by $|\chi_{max,pert}| \sim |\chi|$. This result suggests that the upper bound for the nonperturbative photon-photon interaction will be given by $g_{24}$ in the single-atom case. Consider energies $\hbar(2\omega_{cav} + \epsilon)$ of the $n=2$ manifold eigenstates. In a nonperturbative single-atom regime, there is no evidence that detunings increase the anharmonicity. The limiting case of vanishing detunings and losses gives

$$\epsilon = \pm\sqrt{\frac{G^2}{2} \pm \sqrt{\frac{G^4}{4} - 2g_{13}^2 g_{24}^2}}, \quad (6)$$

where $G^2 = g_{24}^2 + \Omega^2 + 2g_{13}^2$. For the resonant case ($\Delta = \delta = 0$) with $g = g_{13} = g_{24}$, the eigenvectors can be found exactly, and eigenstates whose energy level is closest to $2\hbar\omega_{cav}$ have level shifts that asymptotically approach $g$ from below as $g$ increases. We believe that this result strongly suggests that the maximum possible anharmonicity achievable in a cavity-EIT system is always bounded above by the single atom-cavity coupling coefficient $g_{24}$. When $g_{24} \sim \sqrt{2}g_{13}$ the nonperturbative nature shows itself through level anticrossings. There is then a second upper bound given by $\sqrt{2}g_{13}$.

In order to relate these ideas to well-known photon correlation measurements we calculate $g^2(\tau)$ for experimentally achievable single-atom parameters (Fig. 2) [9]. For weak cw driving such that the average cavity photon number is much less than unity, we expect $g^2(0) \ll 1$ for sufficiently large energy shifts in the $n=2$ manifold. If the coupled system under weak excitation behaves ideally like a single two-level system, then no amplitude for photon modes with $n>1$ will contribute to $g^2(0)$. One can understand this simply as restating the fact that photon blockade implies that, ideally, photons transmitted out of the cavity should only correspond to a cavity decay of $|\phi_0^{n=1}\rangle$. Components from other transitions can be strongly suppressed by large splittings of the dressed states. The main contribution for nonzero $g^2(0)$ would then come from $|\phi_0^{n=2}\rangle$. However, the values we find for $g^2(0)$ are as small as $2\times10^{-3}$ for parameters from recent experiments [10] on cavity-QED ($g=7.5\kappa,\gamma_{31}=0.325\kappa$). Such experiments might be enhanced using EIT. For $\Omega=2.5\gamma_{31}$, so that $\alpha\sim10^2$, one would expect a reduction of the cavity-EIT decay rate from their measured cavity decay rate of 16 MHz to 0.19 MHz. Furthermore, future improvements in cavity and atom trap design will lead to cavity-QED experiments limited by the atomic spontaneous-emission rate; cavity-EIT schemes will be especially favorable in such a case. Clearly the ultralow values of $g^2(0)$ cannot be explained by the level shifts alone. Since the weak probe is tuned exactly between the two dressed states in the $n=2$ manifold, quantum interference might be responsible.

We next discuss how in a high-dispersion regime the usual linear scaling of the nonlinearity with the number of atoms does not apply. The $n=1$ manifold, where a single photon excitation is shared among the $N$ atoms, contains three energy levels $\hbar(\omega_{cav}+\epsilon_i)$ whose energy shifts and widths, neglecting spontaneous emission, are approximately given by Eq. (2), provided we assume that all atoms couple identically to the cavity mode. Note that the eigenvalue $\epsilon_0^{n=1}$ has a width that decreases with the number of atoms $N$ [11], while the splitting of the other two dressed states increases with $N$. The eigenstate corresponding to $\epsilon_0^{n=1}$ is

$$|\phi_0^{n=1}\rangle=\frac{1}{\sqrt{1+\alpha}}\left[\hat{a}^\dagger-\frac{g_{13}}{\Omega}\sum_{i=1}^{N}\hat{\sigma}_{21}^i\right]|0\rangle. \tag{7}$$

When $N>1$, the $n=2$ manifold contains six energy eigenstates (in the perturbative limit). For $g_{24}=0$, one of these eigenstates,

$$|\phi^{n=2}\rangle=\frac{1}{\sqrt{2}(1+\alpha)}\left[\hat{a}^\dagger-\frac{g_{13}}{\Omega}\sum_{i=1}^{N}\hat{\sigma}_{21}^i\right]^2|0\rangle, \tag{8}$$

has energy $\hbar 2\omega_{cav}$, implying that the energy-level diagram, as probed by the weak external field, is harmonic. The state $|\phi^{n=2}\rangle$ corresponds to two cavity-EIT trapping state excitations and has a linewidth that decreases with $1+\alpha$. For $g_{24}\neq0$, this energy eigenstate experiences an ac-Stark shift. When there are no degenerate eigenstates the level shift of $|\phi^{n=2}\rangle$ caused by the perturbation $H_{perturb}$ is

$$\epsilon^{n=2}\approx-2\chi\frac{\alpha}{(1+\alpha)^2}. \tag{9}$$

The atom-cavity molecule therefore has an anharmonic response to the drive field, provided that $|\epsilon^{n=2}|>2\kappa/(1+\alpha)$. We note that the (nonlinear) splitting of the eigenstate corresponding to $\epsilon^{n=2}$ initially increases with the number of atoms $N$ in the cavity, as would be expected from a traditional nonlinear optical system. However, as dispersion becomes important ($\alpha\simeq1$), this increase saturates. In the highly dispersive limit given by $\alpha\gg1$, the splitting of the eigenenergy $\epsilon^{n=2}$ and its width decreases with increasing $N$, under the assumption that all $N$ atoms have the maximum possible interaction.

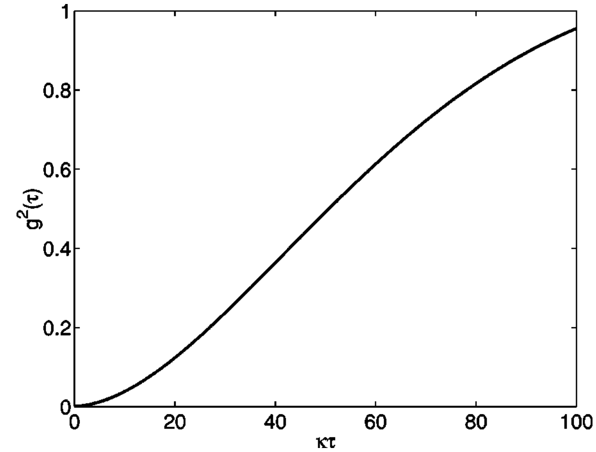


FIG. 2. Second-order photon correlation $g^2(\tau)$ demonstrating photon antibunching. The coupling constant and decay rates correspond to current single-atom experiments [10] of $g_{13}=g_{24}=7.5\kappa$, $\gamma_4=\gamma_{31}=\gamma_{32}=0.325\kappa$, and $\delta=\Delta=0$, $\Omega=2.5\gamma_{31}$, $\mathcal{E}_p=0.1\kappa$.

As we have already argued, the photon-photon interaction strength is determined by the energy-level splittings and linewidths in the $n=2$ manifold, as well as the transition matrix elements. In the special case of doubly resonant EIT ($\delta=0$) with $N>1$, one of the eigenstates

$$|\psi^{n=2}\rangle=\frac{1}{\sqrt{6}}\left[\hat{a}^{\dagger2}+\frac{1}{N}\sum_{i\neq j}\hat{\sigma}_{21}^i\hat{\sigma}_{21}^j+\frac{1}{N}\sum_{i\neq j}\hat{\sigma}_{31}^i\hat{\sigma}_{31}^j\right]|0\rangle \tag{10}$$

has energy $\hbar 2\omega_{cav}$ and a linewidth that is independent of $\alpha$. This state remains unshifted by $g_{24}$ and has a nonzero transition rate from $|\phi^{n=1}\rangle$ that decreases with $\alpha$. Degenerate perturbation theory shows that the actual shifted eigenstate is then a linear combination of $|\psi^{n=2}\rangle$ and $|\phi^{n=2}\rangle$ with level shift $3\epsilon^{n=2}/2$. The transition rate from the $n=1$ manifold EIT state to this shifted eigenstate increases with the number of atoms $N$, for $\alpha>1$, $\chi>\alpha\kappa$. Increasing the number of atoms therefore leads to a reduction in the magnitude of the photon-photon interaction. The presence of these two states implies that the photon blockade is small for this special case of $\delta=0$, where one would naively expect the nonlinearities to be the strongest. It is also interesting to note that the detrimental effect of the degenerate state $|\psi^{n=2}\rangle$ that inhibits nonlinearity implies that the adiabatic elimination of atomic degrees of freedom is not justified, even in the low dispersion limit where it is normally assumed to be valid.

When $|\epsilon^{n=2}|,\kappa<|\delta|<\sqrt{Ng_{13}^2+\Omega^2}$, the degeneracy in the $n=2$ manifold is removed and the dominant contribution for the transition from $n=1$ to $n=2$ manifolds is determined by $|\phi^{n=2}\rangle$. In this case, $P$ becomes independent of $\alpha$ for $\alpha\gg1$ and approaches the single-atom result. Photon blockade experiments using the cavity-EIT system with $N\gg1$ can therefore achieve a reduction in two-photon absorption by the atom+cavity molecule comparable to that of a single atom+cavity. However, the multiatom case is considerably more sensitive to other effects not included in the dressed-state calculations; for example, effective Rabi frequency associated with the external drive has to be much smaller than $\epsilon^{n=2}$, which places a constraint on $\mathcal{E}_p$. Also, the spatial dependence of the atoms, their finite velocities, or atom-atom interactions may become more important. A recent analysis of the spatial dependence of atomic beams [12] for the Jaynes-Cummings model (JCM), where energy levels are proportional to $G_{JCM}=\sqrt{\Sigma_{i=1}^{N}(g_i/g_{max})^2}$, shows that the probability distribution $P(G_{JCM})$ is well approximated by a Gaussian with mean $\sqrt{N_{eff}}$, given by the atomic density and cavity parameters, when $N_{eff}\gg 1$. In the limit $N_{eff}\gg 1$ and $\alpha\gg 1$, the variation in $\epsilon^{n=2}$ level splitting and linewidth over the width of the distribution $P(G_{JCM})$ both decrease like $1/\alpha^2$. The cases $N_{eff}\sim 1$ and $\alpha\sim 1$ are more involved and will be reported elsewhere.

The structure of the eigenstates and energy eigenvalues is the same for any $N>1$, and so we have calculated $g^2(0)$ for $N=2$ versus detuning $\delta$ of the cavity mode from $\omega_{31}$ while maintaining EIT using the coupling constants and decay rates as in the single-atom case (see Fig. 3). The energy-level splittings in the $n=1$ manifold constrain the detuning so that $\delta<\sqrt{g_{13}^2N+\Omega^2}$; we find that large $\delta$ also results in suppression of energy-level splittings in the $n=2$ manifold. As can be seen in Fig. 3, values for $g^2(0)$ using two atoms vary by three orders of magnitude over a detuning $O(g)$; we find that, for such detunings, $g^2(0)\ll 1$, and its value is of the same order of magnitude as that for a single atom for the same driving strength. In contrast, when $\delta=0$, there is practically no photon antibunching for the two-atom case—a result we anticipated based on the appearance of the unshifted eigenstate $|\psi^{n=2}\rangle$. Furthermore, for $|\delta|=g$, $\mathcal{E}_p=0.1\kappa$, $\Omega$ in the range $2\kappa\sim 10\kappa$, we find $g^2(0)\simeq 10^{-3}$. As long as the driving satisfies $\mathcal{E}_p<\kappa/(1+\alpha)$, then large antibunching should be robust. These values for $g^2(0)$ are about two orders of magnitude smaller than for similarly prepared two-level atoms in a cavity.

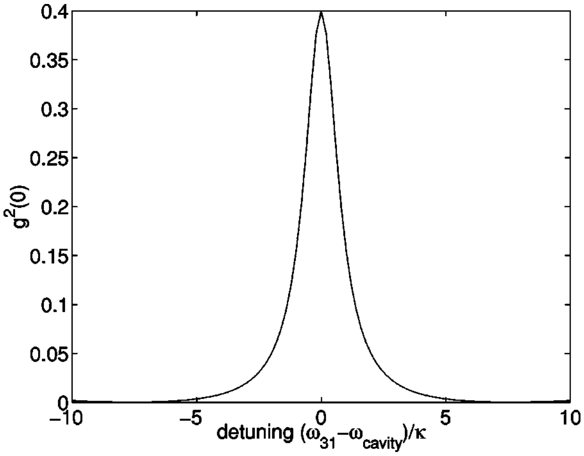


FIG. 3. Two-atom case ($N=2$) showing the variation of $g^2(0)$ with the detuning $\delta=\omega_{31}-\omega_{cav}$ for the cavity-EIT giant Kerr nonlinearity. The coupling constant and decay rates are the same as in Fig. 2, while the detunings and drive strengths are $\Delta=0$, $\mathcal{E}_p=0.01\kappa$.

In summary, we have shown that photon-photon interactions using cavity-EIT are experimentally feasible, and have distinct advantages over two-level atoms. We used the eigenstates of the perturbed many three-level atom system to give the $N$-atom dependence of the energy-level shift. Using an heuristic argument based on transition rates between dressed states we inferred that multiatom photon blockade occurs, provided nonresonant EIT is used. We have shown explicitly that photon antibunching for a multiatom case (two atoms) can be comparable to that of a single atom. We also showed that in the single-atom case, anharmonicity is bounded above by the single atom-photon coupling strength $g_{24}$. This bound is not reached if $\sqrt{2}g_{13}<g_{24}$.

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