Quantum Optical Description of Radiation by a Two-Level System in Strong Laser Fields

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We develop a quantum optical description of radiation from a two-level system (TLS) in strong laser fields, which provides a clear insight into the final states of the TLS and the harmonics field. It is shown that there are two emission channels: the Rayleigh-like channel and the Raman-like channel, which correspond to the TLS ending up in the ground state and excited state after the emission, respectively. The numerical result shows that the harmonics are mainly produced by the Rayleigh-like channel. In addition, according to the coherence of emission among the emitters, the radiation is divided into coherent parts that result from the semi-classical dipole oscillation and incoherent parts that result from the quantum fluctuations of the dipole moment. In the weak field limits, the Rayleigh-like channel corresponds to the coherent parts, and the Raman-like channel corresponds to the incoherent parts. However, in strong laser fields, both channels contribute to coherent and incoherent radiation, and how much they contribute depends on the final excitation. By manipulating the laser field, we can make the Rayleigh-like channel produce either coherent or incoherent radiation.

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Radiation from strong-field-driven atoms has been extensively studied, which covers a wide range of phenomena, including resonance fluorescence and high harmonic generation (HHG).[1–3] These studies have important implications for a variety of applications in fields such as attosecond science, ultrafast optics, and quantum information processing. Recently, quantum optical properties of HHG have received much interest, which is characterized by the non-classical states of light and entanglement between harmonic modes.[4–12] These properties suggest a great potential for realization of HHG-based quantum light sources, which would provide an important platform for quantum information processing.[13]

Currently, radiation is mostly calculated using the semi-classical method, in which the radiation spectrum is typically determined by the time-dependent dipole moment.[14–17] However, limitations of this method have been highlighted by various studies.[4,6,18–21] Especially when one wants to study quantum optical properties of radiation, the quantization of the radiation field must be taken into account. Some studies have attempted to calculate quantum radiation in the laser-dressed picture which enables the separation of coherent and incoherent parts of the radiation.[4] However, these approaches have not yet resolved the final states of the atom and harmonics field.

In this Letter, we investigate the quantum radiation from a laser-driven two-level system (TLS) that can serve as a model for laser-driven quantum wells and solid-state targets.[22,23] While the semi-classical method has been extensively used to study HHG spectra from a laser-driven TLS,[24–26] we aim to develop a quantum optical model that resolves the final states of both the TLS and radiation field. This will allow us to define the emission channel based on the final states of the TLS after the emission and to clarify the coherent contributions of these channels to both the coherent and incoherent radiations.

Theoretical Model. We develop a quantum optical model to describe the radiation from a two-level system driven by a strong laser field. Specifically, we utilize a strong-field Jaynes–Cummings model, in which the driving field is treated as a classical field, while the radiation field is treated as a quantized field. The total Hamiltonian can be expressed as

$$\begin{aligned}\hat{H} &= \hat{H}_{\rm a} + \hat{H}_{\rm f} + \hat{H}_{\rm int}\\ \hat{H}_{\rm a} &= \frac{1}{2}\hbar\omega_{\rm a}\hat{\sigma}_z - \hat{d}E_{\rm L}(t)\\ \hat{H}_{\rm f} &= \sum_{\omega}\hbar\omega\hat{a}_{\omega}^{\dagger}\hat{a}_{\omega},\quad \hat{H}_{\rm int} = -\hat{d}\hat{E}_{\rm R}f(t).\end{aligned} \tag{1}$$

The Hamiltonian $\hat{H}_{\rm a}$ describes a two-level system driven by a classical strong laser field $E_{\rm L}(t)$. The Pauli operator $\hat{\sigma_z}$ is given by $\hat{\sigma_z} = |e\rangle\langle e| - |g\rangle\langle g|$, where the states $|g\rangle$ and $|e\rangle$ denote the eigenstates of the two-level system with excitation energy $\hbar\omega_{\rm a}$. Under the dipole approximation, we express the interaction between the two-level system and the laser field as $-\hat{d}E_{\rm L}(t)$, where the dipole moment operator is given by $\hat{d} = \Omega|e\rangle\langle g| + \Omega^*|g\rangle\langle e|$, and Ω represents the dipole transition matrix element. The Hamiltonian $\hat{H}_{\rm f}$ corresponds to the quantized radiation field, where $\hat{a}_{\omega}$ is the annihilation operator of a photon with frequency ω. The Hamiltonian $\hat{H}_{\rm int}$ describes the interaction between

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the TLS and the quantized radiation field $\hat{E}_{\rm R}$. The field operator $\hat{E}_{\rm R}$ can be expressed as

$$\hat{E}_{\rm R} = -i\sum_{\omega}\sqrt{\frac{\hbar\omega}{2\epsilon_0 V}}(\hat{a}^{\dagger}_{\omega} - \hat{a}_{\omega}), \tag{2}$$

where V is the normalization volume of the quantized fields and ϵ_0 is the vacuum permittivity. The slow-varying temporal function $f(t)$ is introduced to account for the turn-on and turn-off interactions of the laser-driven TLS with the quantized field, and it can be set as the normalized time envelope of the laser pulse.[7]

At the initial time t_0, the TLS is assumed to be in the ground state $|g\rangle$, while the radiation field is assumed to be in the vacuum state $|0\rangle$. Therefore, the initial state of the total system can be represented as $|\psi(t_0)\rangle = |g, 0\rangle$. When the interaction term $\hat{H}_{\rm int}$ is neglected, the dynamics of the laser-driven two-level system and the radiation field can be described using the evolution operator $\hat{U}_0(t, t_0) = \hat{U}_{\rm a}(t, t_0)\hat{U}_{\rm f}(t, t_0)$, where $\hat{U}_{\rm a}(t, t_0)$ and $\hat{U}_{\rm f}(t, t_0)$ correspond to the evolution operators of $\hat{H}_{\rm a}(t)$ and $\hat{H}_{\rm f}$, respectively. The unitary operator $\hat{U}_{\rm a}(t, t_0)$ can be formally expressed as $\hat{U}_{\rm a}(t, t_0) = \hat{T}\int_{t_0}^{t}\exp(-i\hat{H}_{\rm a}(\tau)/\hbar)d\tau$, and it can be non-perturbatively calculated by numerical methods.

Afterwards, the interaction between the TLS and radiation field can be calculated perturbatively. The total evolution operator in the first-order perturbation expansion is given by

$$\hat{U}(t_{\rm f}, t_0) \approx \hat{U}_0(t_{\rm f}, t_0) - \frac{i}{\hbar}\int_{t_0}^{t_{\rm f}}\hat{U}_0(t_{\rm f}, t)\hat{H}_{\rm int}(t)\hat{U}_0(t, t_0)dt. \tag{3}$$

The first term on the right-hand side of Eq. (3) describes the evolution of the TLS and the quantized radiation field without interaction, which implies no emission. The second term introduces the interaction between them at the moment of t, which will lead to the emission. Specifically, when creation operator $\hat{a}^{\dagger}_{\omega}$ is applied to the vacuum state of the harmonic field $|0\rangle$, the harmonic photon will be excited from the harmonics field, i.e., $\hat{a}^{\dagger}_{\omega}|0\rangle = |1_{\omega}\rangle$. Once the harmonic photon is emitted from the TLS, it will not couple again to the TLS because the subsequent couplings are described by the higher-order perturbation terms on $\hat{H}_{\rm int}$, which is negligible in free space due to the extremely small coupling strength.

We can obtain the final state of the system $|\psi(t_{\rm f})\rangle$ by applying the total evolution operator $\hat{U}(t_{\rm f}, t_0)$ to the initial state $|\psi(t_0)\rangle$, which can be expressed as

$$\begin{aligned}|\psi(t_{\rm f})\rangle =& \hat{U}_0(t_{\rm f}, t_0)|g, 0\rangle + \sum_{\omega} g(\omega)e^{-i\omega t_{\rm f}}(d_{\bar{g},g}(\omega)|g, 1_{\omega}\rangle \\ &+ d_{\bar{e},g}(\omega)|e, 1_{\omega}\rangle),\end{aligned} \tag{4}$$

where $g(\omega) = \sqrt{\omega/(2\hbar\epsilon_0 V)}$ represents the coupling strength between the TLS and the quantized radiation field, and $|1_{\omega}\rangle$ denotes the single-photon Fock state. The final state $|\phi, 1_{\omega}\rangle$ indicates the TLS ending up in the final state $|\phi\rangle$ after emitting a photon of frequency ω, where $|\phi\rangle$ denotes either $|g\rangle$ or $|e\rangle$. The probability amplitude is proportional to $d_{\phi,g}(\omega)$, which is defined as

$$d_{\bar{\phi},g}(\omega) = \int_{t_0}^{t_{\rm f}}\langle\phi|\hat{U}_{\rm a}(t_{\rm f}, t)f(t)\hat{d}\hat{U}_{\rm a}(t, t_0)|g\rangle e^{i\omega t}dt. \tag{5}$$

In order to compute $d_{\bar{\phi},g}(\omega)$, it is necessary to calculate the evolution of the initial state $|g\rangle$ from the initial time t_0 to the emission time t, as well as the inverse evolution of the final state $|\phi\rangle$ from the final time $t_{\rm f}$ back to the emission time t. Therefore, the radiation spectrum can be expressed as

$$I(\omega) = \hbar\omega g(\omega)^2\left(|d_{\bar{g},g}(\omega)|^2 + |d_{\bar{e},g}(\omega)|^2\right), \tag{6}$$

where the two terms inside the brackets correspond to the two possible radiation channels resulting in the TLS ending up in either the ground state $|g\rangle$ or the excited state $|e\rangle$ after emitting a photon. In the following context, we will refer to the emission channel where the TLS eventually returns to the ground state as the Rayleigh-like channel. Similarly, the emission channel where the TLS is excited finally will be referred to as the Raman-like channel.

To establish a connection with the semi-classical theory and other quantum optical models on radiation in strong laser fields, we perform a unitary transformation for Eq. (4) by the unitary operator $\hat{U}^{\dagger}_0(t_{\rm f}, t_0)$:

$$\begin{aligned}\hat{U}^{\dagger}_0(t_{\rm f}, t_0)|\psi(t_{\rm f})\rangle =& |g, 0\rangle + \sum_{\omega} g(\omega)(d_{g,g}(\omega)|g, 1_{\omega}\rangle \\ &+ d_{e,g}(\omega)|e, 1_{\omega}\rangle),\end{aligned} \tag{7}$$

where $d_{\phi,g}(\omega)$ is defined as

$$d_{\phi,g}(\omega) = \int_{t_0}^{t_{\rm f}}\langle\phi_i|\hat{U}^{\dagger}_{\rm a}(t, t_0)f(t)\hat{d}\hat{U}_{\rm a}(t, t_0)|\phi_0\rangle e^{i\omega t}dt. \tag{8}$$

Note that the inverse evolution operator $\hat{U}^{\dagger}_{\rm a}(t_{\rm f}, t)$ in Eq. (5) has been replaced with $\hat{U}^{\dagger}_{\rm a}(t, t_0)$ in the above definition. In fact, Eq. (7) represents the final state in the interaction picture with respect to $\hat{H}_0(t)$. Therefore, the radiation spectrum is given by

$$I(\omega) = \hbar\omega g(\omega)^2\left(|d_{g,g}(\omega)|^2 + |d_{e,g}(\omega)|^2\right). \tag{9}$$

Note that the term $|d_{g,g}(\omega)|^2$ now is the Fourier spectrum of the time-dependent dipole moment expectation, which corresponds to the semi-classical result. The other term $|d_{e,g}(\omega)|^2$ is not included in the semi-classical result and can be regarded as the quantum correction, which results from the quantum fluctuation of the dipole moment operator. In addition, for the ensemble of emitters, the radiation from the dipole moment expectation is coherent among the emitters, whereas the radiation from the dipole moment fluctuation is incoherent among the emitters.[4] Therefore, the two terms in Eq. (9) correspond to the coherent and incoherent parts of the radiation, respectively.

So far, we have obtained two formulas for the radiation spectrum, where Eq. (6) can resolve the final state of the TLS and Eq. (9) can distinguish the coherent and incoherent parts of the radiation. Afterward, we will discuss their relation. The unitary transformation presented in Eq. (7) yields

$$\begin{aligned}d^{\rm coh}_{g,g}(\omega) &= d_{\bar{g},g}(\omega)\langle g(t_{\rm f})|g\rangle + d_{\bar{e},g}(\omega)\langle g(t_{\rm f})|e\rangle, \\ d^{\rm incoh}_{e,g}(\omega) &= d_{\bar{g},g}(\omega)\langle e(t_{\rm f})|g\rangle + d_{\bar{e},g}(\omega)\langle e(t_{\rm f})|e\rangle.\end{aligned} \tag{10}$$

Here, $|\phi(t_{\rm f})\rangle$ is defined as $|\phi(t_{\rm f})\rangle = \hat{U}_{\rm a}(t_{\rm f}, t_0)|\phi\rangle$, and $\langle\phi|\phi'(t_{\rm f})\rangle$ denotes the final transition amplitude from the state $|\phi'\rangle$ to $|\phi\rangle$. It is shown that the coherent and incoherent radiation are coherently contributed by both the Rayleigh-like channel $d_{\bar{g},g}(\omega)$ and Raman-like channel $d_{\bar{e},g}(\omega)$. The contribution ratio of the Rayleigh-like and Raman-like channels to the coherent or incoherent radiation is influenced by the transition amplitudes between the eigenstates.

In the weak field limit, the TLS will be adiabatically driven, resulting in $\langle|\phi|\phi'(t_{\rm f})\rangle| = \delta_{\phi,\phi'}$. This yields $|d^{\rm coh}_{g,g}(\omega)|^2 = |d_{\bar{g},g}(\omega)|^2$ and $|d^{\rm incoh}_{e,g}(\omega)|^2 = |d_{\bar{e},g}(\omega)|^2$, and they indicate that the coherent radiation is from the Rayleigh-like channel and the incoherent radiation is from the Raman-like channel. However, in the strong laser field, the final state of TLS is typically the superposition of the ground state and excited state, which indicates that the coherent and incoherent parts of the radiation are both contributed by the two radiation channels. Since the final transition amplitude of the TLS is very sensitive to laser parameters, such as the peak strength and carrier envelop phase, the relative contribution of the two channels can be controlled by manipulating the laser pulse. For example, if the TLS is totally excited at the end of the laser pulse, the coherent radiation will be totally contributed by the Raman-like channel, which is the opposite case of the weak field limit.

Numerical Result. We numerically calculate the radiation spectrum from a TLS in strong laser fields. The electric field of the laser pulse is given by

$$E_{\rm L}(t) = E_0 f(t)\sin(\omega_0 t), \tag{11}$$

where ω_0 is the laser frequency, E_0 is the peak field strength, and $f(t) = \sin^2[\omega_0 t/(2N_{\rm c})]$ is the time envelope with the duration of $N_{\rm c}$ cycles. In the numerical calculation, we set $\omega_0 = \omega_{\rm a}/7$ and $N_{\rm c} = 20$.

Figure 1(a) shows the time-dependent excitation for a laser strength of $E_0 = 11.23\omega_0/|\Omega|$. We can see that the TLS is strongly excited during the laser pulse, and some population remains in the excited state at the end of the laser pulse, i.e. $|\langle e|g(t_{\rm f})\rangle|^2 \neq 0$. The radiation from both the Rayleigh-like channel and Raman-like channel is illustrated in Fig. 1(b), revealing a decline in the lower harmonic order and a plateau in the higher harmonic order.

The low-order harmonics highlighted in the subplot exhibits discrete peaks at odd harmonic orders for both the channels. It is interesting to note that the dips are observed at the harmonic order for the Raman-like channel but not for the Rayleigh-like channel. This observation can be related to Raman and Rayleigh scattering. In typical Raman scattering, the frequency of the scattered light shifts due to a change in the atomic state, while the scattered light has the same frequency as the incident light in Rayleigh scattering. When subjected to a strong laser field, the scattered light includes a series of harmonics due to the nonlinear process. As a result, the TLS emits harmonics and eventually returns to the initial ground state via the Rayleigh-like channel. In contrast, in the Raman-like channel, the TLS emits the satellites around the harmonics and eventually ends up in the excited state.

In addition to the low-order harmonics, both the channels also generate high-order harmonics. However, the HHG intensity produced by the Rayleigh-like channel is much stronger than that from the Raman-like channel. This suggests that the HHG is mainly produced by the Rayleigh-like channel. This result from the quantum radiation theory is consistent with the classical three-step model of the HHG. The classical three-step model describes the HHG in an atom through three key steps: the electron is ionized by the strong laser field, it then gains energy through acceleration in the laser field, and finally recombines with the parent ion and emits HHG. The meaning of the three-step model changes when it is applied to multilevel systems with discrete bound states. In this scenario, the electron is first excited to an excited state, then the excited state is strongly driven by the laser field, and finally, the electron recombines to the initial ground state and emits HHG, which is exactly the process described by the Rayleigh-like channel. Therefore, the Rayleigh-like channel is a quantum version of the classical three-step model.

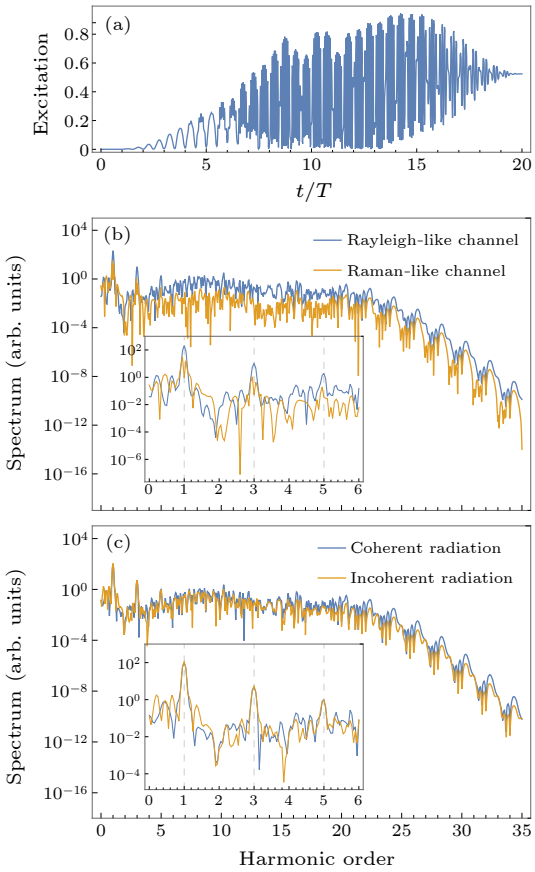

Fig. 1. (a) The time-dependent population in the excited state for a field strength of $E_0 = 11.23\omega_0/|\Omega|$, with T being the laser period. (b) The Fourier spectra $|d_{\bar{g},g}(\omega)|^2$ and $|d_{\bar{e},g}(\omega)|^2$, corresponding to the Rayleigh-like channel and the Raman-like channel, respectively. The insets focus on the low-order harmonics. (c) The Fourier spectra $|d_{g,g}(\omega)|^2$ and $|d_{e,g}(\omega)|^2$, corresponding to the coherent parts and incoherent parts of radiation, respectively.

Figure 1(c) shows the coherent and incoherent harmonics according to Eq. (9). The coherent harmonics re-

sult from the dipole oscillation and represent the semi-classical result, and the incoherent harmonics result from the quantum noise of the dipole moment and represent the quantum correction. Both the coherent and incoherent harmonics are coherently contributed by the Rayleigh-like and Raman-like channels according to Eq. (10). Because the low-order harmonics and HHG is mainly contributed by the Rayleigh-like channel, the intensities of the coherent and incoherent harmonics are determined by the final transition probabilities $|\langle g|g(t_\mathrm{f})\rangle|^2$ and $|\langle e(t_\mathrm{f})|g\rangle|^2$, where they satisfy the relation $|\langle g|g(t_\mathrm{f})\rangle|^2 + |\langle e(t_\mathrm{f})|g\rangle|^2 = 1$. The final transition probability from the ground state to the ground state is $|\langle g|g(t_\mathrm{f})\rangle|^2 = 0.476$, as shown in Fig. 1(a). Therefore, the coherent and incoherent harmonics have nearly equal intensities, as shown in Fig. 1(c).

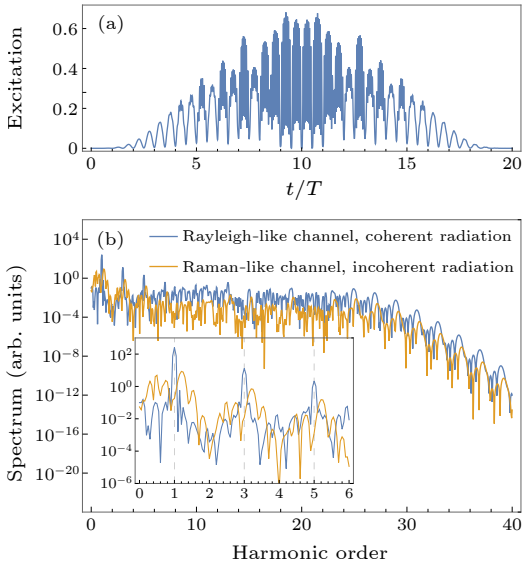

Fig. 2. (a) The time-dependent excitation for a laser strength of $E_0 = 13.21\omega_0/|\Omega|$. (b) The spectrum of coherent radiation completely generated from the Rayleigh-like channel, and the incoherent radiation completely generated from the Raman-like channel.

Since the final transition probabilities between the eigenstates are sensitive to the laser parameter, we can manipulate the relative contributions of two channels to coherent and incoherent radiation. Figures 2 and 3 display two extreme scenarios in which the TLS is completely unexcited and excited, respectively, for laser strengths of $E_0 = 13.21\omega_0/|\Omega|$ and $E_0 = 13.87\omega_0/|\Omega|$. When the TLS is finally in the ground state, the coherent radiation is completely generated from the Rayleigh-like channel, and the incoherent radiation is completely generated from the Raman-like channel, as shown in Fig. 2(b). This leads to the intensity of the coherent HHG stronger than the incoherent HHG. Conversely, when the TLS is completely excited at the end of the laser pulse, the coherent radiation is completely generated from the Raman-like channel, and the incoherent radiation is completely generated from the Rayleigh-like channel, as shown in Fig. 3(b). As a result, the intensity of the incoherent HHG is stronger than that of the coherent HHG, and the coherent low-order harmonics disappear.

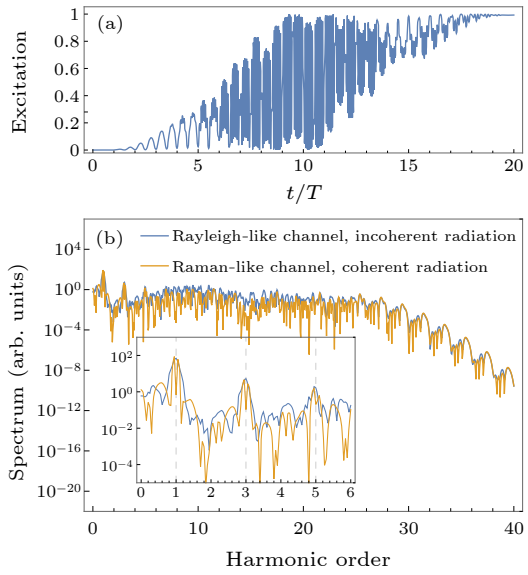

Fig. 3. (a) The time-dependent excitation for a laser strength of $E_0 = 13.87\omega_0/|\Omega|$. (b) The spectrum of coherent radiation completely generated by the Raman-like channel, and the incoherent radiation completely generated by the Rayleigh-like channel.

In summary, we have obtained two formulas, i.e., Eqs. (6) and (9), about the radiation spectrum of the TLS in the strong laser fields by strong-field quantum optical methods. Although the two formulas produce the same total radiation spectrum, they provide different perspectives on the radiation process. Equation (6) defines the Rayleigh-like channel and the Raman-like channel according to the final state of the TLS after the emission. According to the numerical results, the Raman-like channel is responsible for producing the satellites around the low-order harmonics, while the Rayleigh-like channel generates the harmonics themselves. Moreover, the HHG is mainly produced by the Rayleigh-like channel.

Equation (9) divides the radiation into coherent and incoherent parts, which are related to the semi-classical dipole oscillation and quantum fluctuation of the dipole moment, respectively. In the weak field limits, the coherent parts are totally from the Rayleigh-like channel, and the incoherent parts are from the Raman-like channel. However, in the presence of strong laser fields, both the channels contribute to both the coherent and incoherent parts of the radiation, as described by Eq. (10). The relative contribution of each channel is determined by the final transition amplitude between states, which can be manipulated by adjusting the laser pulse.

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