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Frequency-doubled scattering of symmetry-breaking surface-state electrons on liquid helium

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ABSTRACT

Any systems with symmetry-breaking eigenstates can effectively radiate photons with doubled frequency of the incident light, which is known as the second harmonic generation. Here, we study the second-order nonlinear effects with the system of surface-state electrons on liquid helium. Due to the symmetry-breaking eigenstates, we show that an approximate Rabi oscillation between two levels of the surface-state electrons can be realized beyond the usual resonant driving. Consequently, an electromagnetic field with the doubled frequency of the applied driving could be effectively radiated. This can be regarded as a frequency-doubled fluorescence, and interestingly, it works in the unusual Terahertz range.

1. Introduction

An electron (with mass m_e and charge e) near the surface of liquid helium is weakly attracted by its dielectric image potential $V(z) = -\Lambda e^2/z$ [1]; where z is the distance above the helium surface, and $\Lambda = (\varepsilon-1)/4(\varepsilon+1)$, with ε being the dielectric constant of the liquid helium. Due to the Pauli exclusion principle, there is a barrier of about 1 eV for preventing the electron penetrating into liquid helium. Together with such a hard wall at $z=0$, the electron is resulted in an one-dimensional (1D) hydrogen-like spectrum $E_n = -R/n^2$ of motion normal to the Helium surface, with the effective Rydberg energy $R = \Lambda^2 e^4 m_e/(2\hbar^2) \approx 8$ K and Bohr radius $r_b = \hbar^2/(m_e e^2 \Lambda) \approx 76$ Å. These Rydberg levels were first observed by Grimes et al. [2,3] by measuring the resonant frequencies of the transitions between the ground and excited states. They demonstrated a key feature of the surface-state electrons that the resonant frequency can be linearly tuned by a vertical electrostatic field (i.e., the linear Stark effect) due to their symmetry-breaking wave functions. Recently, it has been studied that the above Rydberg states have relatively small line-widths when the temperature of liquid helium is below 1.2 K [4], and thus could be used as qubit (with the ground and first excited states) to realize the interesting quantum information processing, see, e.g., Refs. [5–11].

Here, we first study the second-order nonlinear optical effect of the surface-state electrons on liquid helium. Due to the symmetry-breaking surface-states, the electrons allow an unusual dipole moment which is related to the nonzero average distances between the Rydberg states, i.e., $d = \langle m|z|m\rangle - \langle n|z|n\rangle \neq 0$. Basically, under the resonant driving the usual transition $\langle n|z|m\rangle$ is dominant and the effects related to d are negligible. Alternatively, under the certain large-detuning driving, the effects related to d become important. We show that an approximate Rabi oscillation between the two lowest levels (with the transition frequency ω_e) of the surface-state electrons could be realized by the driving with frequency $\omega_l = \omega_e/2$. This Rabi oscillation leads to an electromagnetic wave emission with the doubled frequency of the applied driving.

2. Rabi oscillations beyond the resonant driving

We consider applying a microwave field $E_l = E_0 \cos(\omega_l t - kx + \phi)$ to a single surface-state electron, where E_0, ω_l, k, and ϕ are its amplitude (in the z-direction), frequency, wave vector, and initial phase, respectively. For simplicity, we consider only two levels (i.e., the ground state $|1\rangle$ and the first excited state $|2\rangle$) of the surface-state electron. Also, we assume that the electron is laterally confined as a quantum dot [5]. Therefore, under the usual dipole approximation $(kx=0)$ the Hamiltonian describing the driven two-level electron

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reads

$$\hat{H}(t) = E_1|1\rangle\langle 1| + E_2|2\rangle\langle 2| - e\hat{z}E_0 \cos(\omega_l t + \phi). \quad (1)$$

By using the completeness relation $|1\rangle\langle 1| + |2\rangle\langle 2| = \hat{\mathbf{1}}$, one can write $\hat{z}$ as

$$\hat{z} = z_{11}|1\rangle\langle 1| + z_{22}|2\rangle\langle 2| + z_{12}|1\rangle\langle 2| + z_{21}|2\rangle\langle 1|, \quad (2)$$

with the dipole matrix elements $z_{12} = \langle 1|\hat{z}|2\rangle = \langle 2|\hat{z}|1\rangle = z_{21}$. Due to the asymmetry eigenstates $|1\rangle$ and $|2\rangle$, here $z_{11} = \langle 1|\hat{z}|1\rangle$ and $z_{22} = \langle 2|\hat{z}|2\rangle \neq 0$. This is very different from the case for the natural atoms wherein $z_{11} = z_{22} = 0$. By using Eq. (2) we can rewrite Hamiltonian (1) as

$$\hat{H}(t) = \frac{\hbar\omega_e}{2}\hat{\sigma}_z - 2[\hbar\tilde{\Omega}\hat{\sigma}_z + \hbar\Omega_R(\hat{\sigma}_{12} + \hat{\sigma}_{21})] \cos(\omega_l t + \phi), \quad (3)$$

and further

$$\begin{aligned}\hat{H}(t) = &-\hbar\tilde{\Omega}\hat{\sigma}_z(e^{i\omega_l t + i\phi} + \text{H.c.}) \\ &-\hbar\Omega_R[\hat{\sigma}_{12}(e^{-i\Delta t + i\phi} + e^{-i(\omega_e + \omega_l)t - i\phi}) + \text{H.c.}],\end{aligned} \quad (4)$$

in the interaction picture defined by $\hat{V} = \exp(-it\omega_e\hat{\sigma}_z/2)$. Above, $\hat{\sigma}_z = |2\rangle\langle 2| - |1\rangle\langle 1|$ is the Pauli operator with the transition frequency $\omega_e = (E_2 - E_1)/\hbar$, $\hat{\sigma}_{ij} = |i\rangle\langle j|$ is the projection operator (with $i, j = 1, 2$), $\Delta = \omega_e - \omega_l$ is the detuning between the microwave and the electron, $\Omega_R = ez_{12}E_0/(2\hbar)$ is the usual Rabi frequency, and finally $\tilde{\Omega} = e(z_{22} - z_{11})E_0/(4\hbar) \neq 0$ due to the symmetry-breaking in the states $|1\rangle$ and $|2\rangle$.

Under the resonant condition, i.e., $\Delta = 0$, the evolution ruled by the Hamiltonian (4) can be approximately expressed as

$$\hat{U}(t) = 1 + \left(\frac{-i}{\hbar}\right)\int_0^t \hat{H}(t_1)\, dt_1 + \cdots \approx \hat{T} \exp\left[\int_0^t \hat{H}_R(t)\, dt\right] \quad (5)$$

with the Dyson-series operator $\hat{T}$ and the effective Hamiltonian:

$$\hat{H}_R = -\hbar\Omega_R(e^{i\phi}\hat{\sigma}_{12} + e^{-i\phi}\hat{\sigma}_{21}). \quad (6)$$

Certainly, this Hamiltonian describes the standard Rabi oscillation with frequency Ω_R. Above, the terms related to $\xi = (\Omega_R, \tilde{\Omega})/\omega_e$ are neglected, since $\xi \ll 1$ under the weak drivings. In fact, this approximation is nothing but the usual rotating-wave approximation. Indeed, under the resonant excitation, the contribution from the symmetry-breaking $\hbar\tilde{\Omega}\hat{\sigma}_z$ is negligible, due to its small probability proportional to ξ.

Interestingly, under the large-detuning driving $\Delta = \omega_l$, i.e., $\omega_l = \omega_e/2$, the effects related to the symmetry-breaking $\hbar\tilde{\Omega}\hat{\sigma}_z$ become significant. This can be seen from the following new evolution operator [12]:

$$\begin{aligned}\hat{U}(t) = &1 + \left(\frac{-i}{\hbar}\right)\int_0^t \hat{H}(t_1)\, dt_1 \\ &+ \left(\frac{-i}{\hbar}\right)^2 \int_0^t \hat{H}(t_1)\int_0^{t_1} \hat{H}(t_2)\, dt_2\, dt_1 + \cdots \\ \approx &\hat{T} \exp\left[\int_0^t \hat{H}_L(t)\, dt\right],\end{aligned} \quad (7)$$

with the second-order effective Hamiltonian

$$\hat{H}_L = \frac{\hbar\delta}{2}\hat{\sigma}_z - \hbar\Omega_L(e^{i2\phi}\hat{\sigma}_{12} + e^{-i2\phi}\hat{\sigma}_{21}) \quad (8)$$

and where $\delta = 4\Omega_R^2/\omega_e$. Due to the weak driving $(\Omega_R, \tilde{\Omega}) \ll (\omega_l, \omega_e)$, the present Rabi frequency $\Omega_L = 4\Omega_R\tilde{\Omega}/\omega_e$ is smaller than the previous Ω_R for the resonant excitation. Also, all the effects related to the small quantity $\xi = (\Omega_R, \tilde{\Omega})/(\omega_l, \omega_e)$ were neglected, but the terms containing $\Omega_R\tilde{\Omega}/(\omega_l, \omega_e)$ were retained. Note that, under the above large-detuning driving, the usual first-order expansion term $\hat{U}^{(1)}(t) = (-i/\hbar)\int_0^t \hat{H}(t_1)\, dt_1$ practically does not contribute to the time evolution, due to its small probability: $P^{(1)}(t) \sim \xi$.

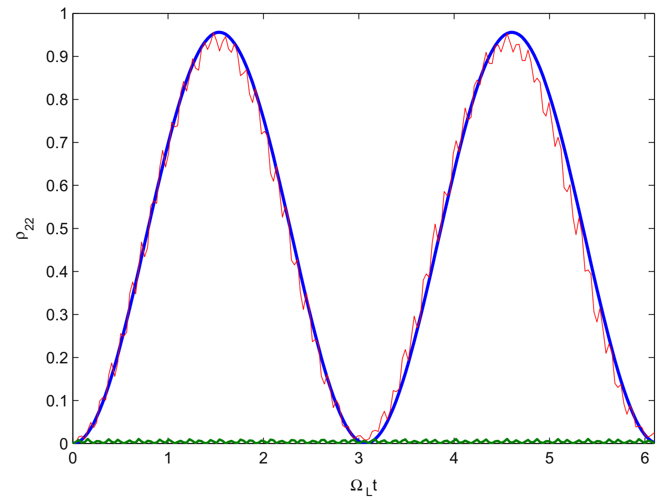

Fig. 1. The probability ρ_{22} of excited state $|2\rangle$ versus $\Omega_L t$. The blue and red lines correspond respectively to the analytical solution from the second-order effective Hamiltonian (8) and the numerical one from the original Hamiltonian (3). Assuming $\tilde{\Omega} = 0$, similar to that of the usual atoms, the time dependent ρ_{22} is given by the green line. These lines show that the effect due to the symmetry-breaking $\tilde{\Omega} \neq 0$ is significant under large-detuning driving $\omega_l = \omega_e/2$. (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this paper.)

Because of the term $\hbar\delta\hat{\sigma}_z/2$ (corresponding to the usual ac-stark effect [13]), the Hamiltonian (8) is not the standard Rabi model. However, it could be regarded as an approximate one when $\delta/\Omega_L < 1$. This can be seen from Fig. 1, where the time dependent ρ_{22} is plotted. The relevant parameters are selected as the typical [3,6]: $\omega_e \approx 0.22$ THz, $z_{12} \approx 0.5\, r_b$, and $z_{22} - z_{11} \approx 2.3 r_b$. Consequently $\delta/\Omega_L = 2z_{12}/(z_{22} - z_{11}) \approx 0.43$, $\Omega_R \approx 4.3$ GHz, and $\Omega_L \approx 0.78$ GHz with the microwave amplitude $E_0 \approx 15$ V/cm. Fig. 1, with numerical solutions from the Hamiltonian (3), proves that the above derivation for effective Hamiltonian (8) is valid and the effect due to the symmetry-breaking $\tilde{\Omega} \neq 0$ is significant indeed.

3. Frequency-doubling radiations

We now consider the steady-state radiations due to the above oscillations of occupancies by taking into account the practically existing dissipations. For this case the dynamics of the driven surface-state electrons is described by the master equation

$$\begin{aligned}\frac{d}{dt}\hat{\rho} = &\frac{-i}{\hbar}[\hat{H}_L, \hat{\rho}] + \frac{\Gamma}{2}(2\hat{\sigma}_{12}\hat{\rho}\hat{\sigma}_{21} - \hat{\sigma}_{21}\hat{\sigma}_{12}\hat{\rho} - \hat{\rho}\hat{\sigma}_{21}\hat{\sigma}_{12}) \\ &+ \frac{\gamma}{2}(2\hat{\sigma}_{22}\hat{\rho}\hat{\sigma}_{22} - \hat{\sigma}_{22}\hat{\rho} - \hat{\rho}\hat{\sigma}_{22})\end{aligned} \quad (9)$$

with Γ and γ being the decay and dephasing rates, respectively. $\hat{\rho} = \sum_{i,j=1}^2 \rho_{ij}|i\rangle\langle j|$ is the density operator of the two-level quantum system, and the matrix elements $\{\rho_{ij}\}$ obey the normalized and hermitian conditions: $\sum_{i=1}^2 \rho_{ii} = 1$ and $\rho_{ij} = \rho_{ji}^*$, respectively. In the two-level atomic representation, the above master equation takes the following matrix forms:

$$\begin{cases}\dfrac{d}{dt}\rho_{22} = i\Omega_L(\rho_{12}e^{-i\phi} - \rho_{21}e^{i\phi}) - \Gamma\rho_{22}, \\ \dfrac{d}{dt}\rho_{21} = i(\rho_{11}\Omega_L e^{-i\phi} - \rho_{22}\Omega_L e^{-i\phi} - \delta\rho_{21}) - \dfrac{\Gamma + \gamma}{2}\rho_{21}.\end{cases} \quad (10)$$

Certainly, due to the resistance from the surroundings, the amplitudes of oscillating ρ_{ii} decrease with the time evolving (see Fig. 2), and $d\rho_{ii}/dt \to 0$ when $t \to \infty$. Specially, under the steady-state