


## Image-Charge Detection of the Rydberg States of Surface Electrons on Liquid Helium

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We propose and experimentally demonstrate a new spectroscopic method, image-charge detection, for the Rydberg states of surface electrons on liquid helium. The excitation of the Rydberg states of the electrons induces an image current in the circuit to which the electrons are capacitively coupled. In contrast to the conventional microwave absorption measurement, this method makes it possible to resolve the transitions to high-lying Rydberg states of the surface electrons. We also show that this method can potentially be used to detect quantum states of a single electron, which would pave a way to utilize the quantum states of the surface electrons on liquid helium for quantum computing.

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Surface electrons (SE) above liquid helium constitute an exquisitely pure quantum system which for a long time served as a unique experimental platform to discover interesting many-electron phenomena [1]. The quantized (Rydberg) states of SE with a hydrogenlike energy spectrum  $E_n = -R_e/n^2$ , where  $R_e \sim 10^{-3}$  eV and  $n$  is a positive integer number, are formed due to the attractive interaction between an electron and its image charge inside the liquid, as well as a strong repulsive barrier experienced by the electron at the vapor-liquid interface. For typical experimental temperatures below 1 K, electrons occupy the ground state and are localized about 10 nm above the surface. The higher-energy Rydberg states can be excited by millimeter waves radiation. Grimes and Brown first measured the transitions from the ground ( $n = 1$ ) to the low-lying excited states ( $n = 2 \sim 6$ ) by detecting the change in the microwave (MW) absorption caused by the excitation of the Rydberg states of SE using a cryogenic bolometer [2,3]. A renewed interest in the Rydberg states of SE has emerged from their potential as qubit states [4,5], which was followed by several other proposals to use either orbital or spin states of SE as qubit states [6,7]. A crucial point for successful qubit implementation is the ability to manipulate and detect quantum states of a single electron. For such an application, the conventional MW absorption measurement is inappropriate for a detection of the quantum state because it is applicable only for a sufficiently large number of electrons [2,8,9], and so is the indirect detection of the Rydberg transitions via the measurement of SE conductivity [10,11]. Originally, a destructive readout of the Rydberg states was proposed, in which the electrons leave the liquid surface depending on the occupied Rydberg states [4,12]. An interesting and promising idea is to use the strong coupling of a single electron to a superconducting resonator to realize a nondestructive readout of electron quantum states [7,13]. However, this

method is limited to a low transition frequency which should match the frequency of the coplanar resonator ( $\sim 5$  GHz); thus it is not applicable for the detection of the excitation of the Rydberg states.

In this Letter, we propose and demonstrate a new spectroscopic method, image-charge detection, for the Rydberg states of SE on liquid helium. Our method makes use of the fact that, as an electron is excited from the ground state to a higher excited state, the electron wave function spreads farther away from the liquid surface and its average distance from the liquid surface increases. Thus, when the system is placed near an electrode aligned parallel to the liquid surface, the excitation of SE causes a change in the image charge induced in the electrode by SE. In our experiment, the method is demonstrated with MW-excited SE placed in a parallel-plate capacitor. By measuring the image current in the capacitor, we managed to detect the excitation to the high-lying Rydberg states which were unable to be measured with the above-mentioned conventional methods. We also discuss an alternative detection scheme which can be scaled down to detection of the excitation of a single electron.

The principle diagram of the image-charge detection is shown in Fig. 1. SE (light blue circles) are formed on the surface of liquid helium which are placed between two plates of a capacitor. Here, we use a parallel-plate capacitor (the area of each plate  $S$  and the distance between the plates  $D$ ). SE are confined on the surface of liquid helium by applying a positive dc bias  $V_{dc}$  to the bottom capacitor plate. Owing to the linear Stark shift of the Rydberg levels caused by a dc electric field  $E_{\perp}$  applied perpendicular to the surface, the transition frequency of SE,  $\omega_{1n}(E_{\perp}) = (E_n - E_1)/\hbar$ , can be adjusted to match with the microwave frequency  $\omega_0$  by varying the value of  $V_{dc}$ . The MW-excited electrons (green circles in Fig. 1) are elevated above the charged layer of the ground-state electrons by a distance

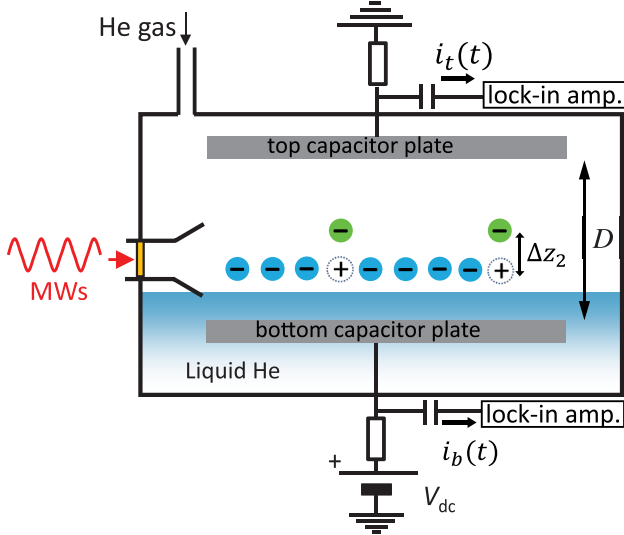


FIG. 1. Principle diagram of the image-charge detection method (see text for details).

$\Delta z_n = z_{nn} - z_{11}$ , where  $z_{nn}$  is the average value of the  $z$  coordinate (counted from the surface) of an electron occupying the Rydberg state of index  $n$ . For the first excited state,  $n = 2$ , this distance is  $\Delta z_2 \approx 35$  nm for  $E_{\perp} = 0$ , and somewhat decreases with increasing  $E_{\perp}$  [5]. As a fraction  $\rho_{nn}$  of SE is excited to the  $n$ th Rydberg state, the image charge induced by SE in the top (bottom) capacitance plate changes by  $q$  ( $-q$ ):

$$q = \frac{\Delta z_n}{D} e n_s \rho_{nn} S = \frac{\Delta z_n}{D} P_e S, \quad (1)$$

where  $e(> 0)$  is the elementary charge and  $n_s$  is the areal density of SE. Here, for later discussion, we introduce the quantity  $P_e = n_s S \rho_{nn} p / (S \Delta z_n) = e n_s \rho_{nn}$ , with  $p = e \Delta z_n$  being the electric dipole moment of one electron. This quantity can be viewed as the electric dipole moment per unit volume of the electron system induced by the excitation of SE to the  $n$ th excited state. The change in the image charge induces a current  $i_t$  in the top plate (and a current  $i_b$  of the opposite sign in the bottom plate), which is given by the time derivative of Eq. (1). These currents can be detected using a lock-in amplifier by periodically varying the fractional occupancy  $\rho_{nn}$ , and therefore the electrical dipole moment  $P_e$  of SE. In the experiment described below, this is done by applying a pulse-modulated MW excitation to the system. Assuming the harmonic time dependence of  $\rho_{nn}$  at the pulse modulation frequency  $\omega_m$ :  $\rho_{nn} = \rho_{nn}^{(0)} e^{i\omega_m t}$ , the current amplitude can be estimated as

$$|i_{t,b}(t)| = \left| \frac{dq}{dt} \right| = \frac{e n_s C_0 \omega_m \Delta z_n \rho_{nn}^{(0)}}{\epsilon_0}, \quad (2)$$

where  $C_0 = \epsilon_0 S / D$  is the capacitance of the parallel plate capacitor and  $\epsilon_0$  is the vacuum permittivity. For typical

values of  $n_s = 10^8$  cm $^{-2}$ ,  $C_0 \sim 1$  pF, the modulation frequency  $\omega_m / 2\pi = 100$  kHz, and  $\rho_{nn}^{(0)} = 10\%$ , we obtain  $|i_{t,b}(t)| \sim 100$  pA.

The experiment is carried out in a leak-tight copper cell cooled to  $T = 0.2$  K in a dilution refrigerator and filled with condensed  $^3\text{He}$  gas. The helium liquid surface is set approximately midway between two plates of a parallel-plate capacitor formed by two round (diameter 24 mm) conducting plates separated by four quartz spacers of height 2 mm, which sets the gap  $D$  between the capacitor plates. Electrons are produced by the thermionic emission from a tungsten filament placed near the capacitor and above the liquid surface, and electrons are attracted towards the liquid surface by applying a positive bias  $V_{dc}$  to the bottom capacitor plate (see Fig. 1). Electrons charge the surface of liquid above the biased plate until they completely shield the electric field above the surface. After charging the surface, the filament is turned off and the saturated density of electrons remains constant during the experiments.

MW radiation at a fixed frequency  $\omega_0$  is introduced into the cell through a sealed rectangular single-mode waveguide with inner dimensions  $1.6 \times 0.8$  mm. In order to increase the area illuminated by MW, the waveguide gradually transforms to an overmoded size ( $3.8 \times 1.9$  mm) inside the cell (shown schematically in Fig. 1). A resonant  $1 \rightarrow n$  transition between the Rydberg states of SE is excited by adjusting voltage  $V_{dc}$  to match  $\omega_{1n}$  with  $\omega_0$  via Stark shift. In the experiment, we employ on/off MW pulse modulation at frequency  $\omega_m$  and measure the ac currents at either the bottom or top capacitor plates using a lock-in amplifier, while sweeping  $V_{dc}$  through the resonance. The typical result of such measurements is shown in Fig. 2 where

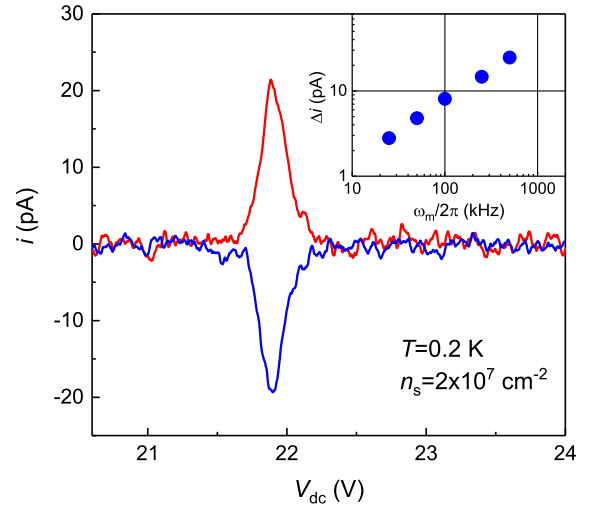


FIG. 2. The current signal measured at the bottom (red line) and top (blue line) capacitor plate for SE irradiated with pulse-modulated MW at frequency  $\omega_0 / 2\pi = 140$  GHz. (Inset) The current signal at the resonance measured for different values of the pulse-modulation frequency  $\omega_m$ .

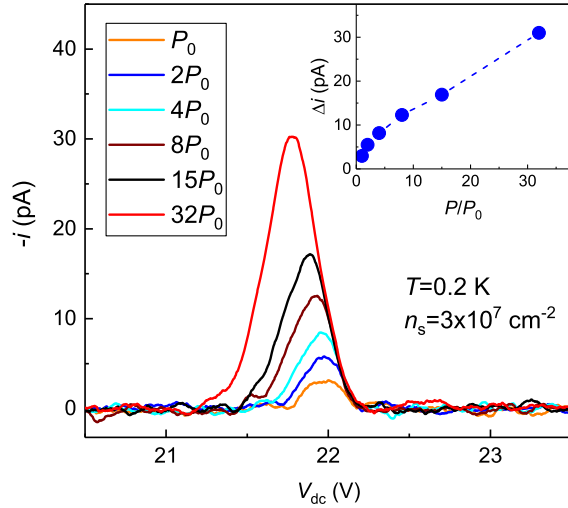


FIG. 3. The current signal measured at the top capacitor plate for SE irradiated with pulse-modulated MW at frequency  $\omega_0/2\pi = 140$  GHz for different MW power. Inset: The current signal at the resonance vs the normalized MW power.

we plot the in-phase component of the current measured by the lock-in amplifier at the modulation frequency  $\omega_m/2\pi = 250$  kHz. The sharp enhancement of current corresponds to the resonant  $1 \rightarrow 2$  transition of SE excited by the applied MW at frequency  $\omega_0/2\pi = 140$  GHz. The resonance value of  $V_{dc}$  corresponds to the electric field acting on the electrons  $E_{\perp} = V_{dc}/D \approx 110$  V/cm. To compare this result with the theoretical prediction, we numerically calculated the Rydberg energy spectrum of an electron above liquid  $^3\text{He}$  for different values of  $E_{\perp}$  assuming an infinitely large potential barrier for electrons at the vapor-liquid interface. For  $\omega_{12}/2\pi = 140$  GHz, we obtained  $E_{\perp} = 116$  V/cm which agrees reasonably well with the measured value. The deviation of the experimental result from the prediction of our theoretical model is due to finiteness of the repulsive barrier [3], which is equal to approximately 1 eV.

At  $T = 0.2$  K, the transition linewidth for an electron bound to the surface of liquid  $^3\text{He}$  is determined by its scattering from surface capillary waves (ripples). The full width at half-maximum of the transition peak in Fig. 2 is about 400 MHz, which is significantly larger than the expected intrinsic linewidth  $\gamma \approx 10$  MHz due to the scattering from ripples [9,14]. This width is determined by the inhomogeneous broadening of the transition energy due to the nonuniformity of  $E_{\perp}$ . An important feature of the result shown in Fig. 2 is that the current signals measured at the top and bottom plates are nearly equal in magnitude but opposite in sign, as expected. The inset of Fig. 2 shows a nearly linear dependence of the magnitude of  $i$  at the resonance for different values of  $\omega_m$ , as expected from Eq. (2).

Figure 3 shows the dependence on MW power of the current signal measured at the top plate for the resonant

$1 \rightarrow 2$  transition. The magnitude of the current at the resonance vs normalized power is shown in the inset of this figure. For a two-level system, the fractional occupancy  $\rho_{22}$  is expected to increase linearly for small powers and saturate at the value 0.5 at high powers. The measured dependence shows a deviation from such a dependence. It is known that in SE the two-level approximation fails for a high MW power due to the electron heating by MW radiation, which results in the thermal population of the Rydberg states that are not directly excited by MW [11]. Thus, the expression  $\Delta z_n \rho_{nn}^{(0)}$  in Eq. (2) becomes reformulated as  $\sum_{k \geq n} \Delta z_k \rho_{kk}^{(0)}$ . The data in Fig. 3 show that the resonance shifts towards lower values of  $V_{dc}$  with increasing power. This is due to the Coulomb shift of the Rydberg levels caused by the electron-electron interaction [15]. Using the experimentally determined  $d\omega_{12}/dV_{dc} \approx 2.1$  GHz/V, the estimated Coulomb shift is about 0.2 GHz for the highest applied MW power (red line in Fig. 3). Assuming the Boltzmann population for all the Rydberg states, the corresponding estimated electron temperature is  $T_e \approx 5$  K [11].

Figure 4 shows the current signal measured at a fixed MW power as  $V_{dc}$  is swept to zero, i.e., tuning transitions to higher- $n$  states to the resonance with the applied MW. We can clearly observe the transitions up to  $n = 14$ , while the transitions to high-lying states were difficult to resolve with conventional MW absorption measurements [3]. As the Thomas-Reiche-Kuhn sum rule states that  $\sum_{n=1}^{\infty} \hbar \omega_{1n} |\langle 1|z|n \rangle|^2$  converges, the absorbed

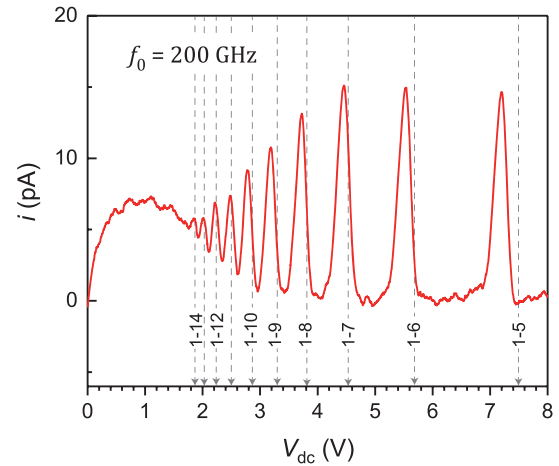


FIG. 4. The current signal measured at the bottom capacitor plate for SE irradiated with pulse-modulated MW at frequency  $\omega_0/2\pi = 200$  GHz vs bias  $V_{dc}$  applied to the bottom electrode. The dashed lines indicate theoretical predictions for the transitions between the ground state and excited states using the approximate model with infinitely large potential barrier [3]. The deviation from the theoretical model is due to the finiteness of the barrier and decreases with increasing  $n$  because an electron occupying high-lying Rydberg states is less sensitive to the potential barrier at the surface.

MW power, which is proportional to the transition rate, decreases rapidly with increasing  $n$ . In the case of the image current [Eq. (2)], the decrease in the transition rate is compensated by the increase in  $\Delta z_n$ . At lower  $V_{dc}$ , since the Stark shift of the Rydberg levels is smaller, the overlap between different transitions becomes larger and produces a smooth background, which makes it difficult to resolve each transition.

Next, we discuss the potential improvements of the demonstrated method towards detection of the Rydberg state excitation of a single electron. In particular, we would like to point out the similarity between our image-charge detection and single-electron capacitance spectroscopy in semiconductor quantum dots [16]. Let us apply a small ac voltage  $u(t)$  to the plate of capacitor in Fig. 1 in addition to the dc bias  $V_{dc}$  to induce the periodic variation of the fractional occupancy  $\rho_{nn}$ . We introduce the electric field  $\mathcal{E} = u/D$  due to the ac voltage applied to the capacitor and expand  $\rho_{nn}$  in the Taylor series as  $\rho_{nn}(u) = \rho_{nn}|_{u=0} + (d\rho_{nn}/d\mathcal{E})|_{u=0}\mathcal{E} + (d^2\rho_{nn}/d\mathcal{E}^2)|_{u=0}(\mathcal{E}^2/2) + \dots$ . Then, the electric dipole moment per unit volume of the electron system can be cast in the form  $P_e = P_e^{(0)} + \epsilon_0\chi_e\mathcal{E}$ , where  $\chi_e = \chi_e^{(1)} + \chi_e^{(2)}\mathcal{E} + \dots$  is the nonlinear ac electric susceptibility of the electron system. By inserting this form into Eq. (1), the ac current in the capacitor can be written as

$$i(t) = \frac{d[C_0u(t) + q]}{dt} = C_0 \left( 1 + \chi_e^{(1)} \frac{\Delta z_n}{D} + \chi_e^{(2)} \frac{\Delta z_n 2u(t)}{D^2} + \dots \right) \frac{du(t)}{dt}. \quad (3)$$

The linear part of susceptibility  $\chi_e^{(1)}$  is given by

$$\chi_e^{(1)} = -\frac{\alpha \epsilon n_s}{\epsilon_0} \left( \frac{\partial \rho_{nn}}{\partial \omega_{1n}} \right), \quad (4)$$

where we used an approximated linear dependence of  $\omega_{1n}$  vs  $E_\perp$  with the slope  $\alpha = d\omega_{1n}/dE_\perp$ . Equation (3) states that the change in the capacitance  $C_0$  due to the linear part of  $\chi_e$  is given by

$$\Delta C = C_0 \chi_e^{(1)} \frac{\Delta z_n}{D} = \frac{\Delta z_n \alpha \epsilon N_e}{D^2} \left( \frac{\partial \rho_{nn}}{\partial \omega_{1n}} \right), \quad (5)$$

where  $N_e$  is the number of surface electrons in the capacitor. This change in capacitance can be regarded as being caused by the electric susceptibility of SE induced by the MW-induced population of the excited states. From another point of view, it has the same physical mechanism as the change in the quantum capacitance due to electron tunneling in quantum dots [17,18]; in both cases it is caused by the change in the image charge at the capacitor plate due to the change in the spatial position of an electron coupled to the capacitor. To estimate the value of  $\Delta C$  for a single

electron,  $N_e = 1$ , we can assume  $\partial \rho_{nn}/\partial \omega_{1n} \sim \gamma^{-1}$ , where  $\gamma$  is the intrinsic linewidth due to scattering from ripplons. Using  $\gamma = 10$  MHz (corresponding to  $T = 0.2$  K) [9,14] and  $\alpha = 5$  MHz/(V/m) obtained from the numerical calculations of the Rydberg spectrum of an electron as were described earlier, for  $D = 2$  mm we obtain  $\Delta C \sim 10^{-3}$  aF. While it is too small a change to be detected, we note that it crucially depends on the capacitance gap  $D$ , see Eq. (5). By employing a capacitor with  $D = 10$   $\mu\text{m}$ , the change in capacitance due to single-electron excitation is  $\sim 10$  aF. This is a typical change in the capacitance due to single-electron tunneling in semiconductor quantum dots, which can be detected by dispersive changes in a resonant rf tank (LC) circuit [17,18]. This makes the image-charge detection of the Rydberg transition of a single electron on liquid helium promising. We note that it is not practical to use a parallel-plate capacitor, having a gap  $D$  less than  $\sim 1$  mm because the liquid helium would completely fill the gap due to the capillary action [19]. Instead, it would be convenient to employ a coplanar capacitor with both plates located in the same plane of a solid substrate and covered with the liquid helium film [20].

Once the detection of the excitation of the Rydberg state is realized for a single electron, we could transform it to a nondestructive readout of the spin state of a single electron with the help of a magnetic field gradient. A current running through a superconducting wire in the vicinity of a trapped electron [7] or a ferromagnet under an external magnetic field [21] can create a local magnetic field gradient in the direction perpendicular to the surface of liquid helium. Thanks to the field gradient, the electron feels a different magnetic field depending on the Rydberg state. The energy difference between the Rydberg-ground state and a Rydberg-excited state becomes also different depending on the spin state. Thus, the microwave frequency required to excite the Rydberg state becomes spin dependent. By applying MW radiation and setting its frequency for the excitation of the spin-up state, the image-charge detection picks up the signal only when the electron is in the spin-up state. The detailed schematics are beyond the scope of this paper and will be discussed elsewhere.

In summary, we have proposed a new method, the image-charge detection, for spectroscopic study of the Rydberg states of surface electrons on liquid helium. The method is demonstrated by measuring the image current induced in the capacitor circuit by a pulse-modulated MW excitation. The method is simple and does not require expensive devices such as a cryogenic bolometer. It is demonstrated that the image-charge detection provides other advantages over the conventional methods, such as the ability to do spectroscopic studies of the high-lying Rydberg states. We also discussed the possibility of using this method to detect the spin state of a single electron, which would open a new pathway for using spin states of SE on liquid helium for quantum computing.



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