Microwave Saturation of the Rydberg States of Electrons on Helium

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We present measurements of the resonant microwave excitation of Rydberg energy levels for surface-state electrons on superfluid helium. The temperature-dependent contribution to the linewidth $\gamma(T)$ agrees with theoretical predictions and is very small below 700 mK, in the ripplon scattering regime. Absorption saturation and power broadening were observed as the fraction of electrons in the first excited state was increased to 0.49, close to the thermal excitation limit of 0.5. The Rabi frequency Ω was determined as a function of microwave power. High values of the ratio Ω/γ confirm this system as an excellent candidate for creating qubits.

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Surface state electrons on liquid helium [1] are attracted by a weak positive image charge in the helium. This Coulomb potential well for vertical motion produces a hydrogenlike spectrum, with energy levels $E_m = -R_e/m^2$ where R_e is the effective Rydberg energy (0.67 meV) and $m(\geq 1)$ is the quantum number, shown schematically in Fig. 1. These Rydberg states were first observed by Grimes *et al.* [2] who measured the resonant frequency f_{1m} and the linewidth $\gamma(T)$ above 1.2 K for transitions from the ground state to the excited states $m = 2, 3$. A key feature is a linear Stark effect, due to asymmetry of the wave functions, so that the resonant frequency can be tuned with a vertical electric field E_z. Their measurements of f_{1m} versus E_z showed excellent agreement with theoretical calculations from $f_{12} = 125.9$ GHz in zero holding field up to 220 GHz for $E_z = 17.5$ kV/m. Lambert and Richards [3] extended the frequency range up to 765 GHz using a far-infrared laser. Edel'man and later Volodin and Edel'man [4] indirectly probed the Rydberg states, on both liquid ^{4}He and liquid ^{3}He, by measuring the photoconductivity (the change in the electron mobility) when the electrons were excited by incident microwaves. They estimated that the total intrinsic linewidth at 0.4 K for electrons on ^{4}He was $\leqslant 15$ MHz, consistent with the theory of Ando [5], with inhomogeneous broadening of the same magnitude.

Interest in these states has now been rekindled by the suggestion of Platzman and Dykman [6] that electrons on helium could be used as electronic qubits, with the ground and the first excited states representing $|0\rangle$ and $|1\rangle$, respectively. The qubits would be controlled using resonant microwave excitation. The potential is anharmonic so that the two lowest states are an excellent approximation to a two-level system with minimal coupling to higher levels. The formalism is similar to nuclear magnetic resonance (NMR), governed by the optical Bloch equations [7]. The linewidth is related to the NMR relaxation rate $1/T_1$ and the decoherence rate $1/T_2$, describing the quantum system [8]. In principle, a surface-state electron quantum computer would be similar to the NMR quantum computers recently developed [9,10]. The potential advantages come from the exceptional properties of the electronic system itself and the scalability of custom-designed qubits. The results presented here were obtained as part of an experimental program to develop such a system [11].

In particular, we report the first direct measurements below 1.2 K of the microwave absorption between the Rydberg states of surface-state electrons on helium. Two key aspects for qubits are the linewidth of the resonant microwave absorption, which must be small to limit decoherence, and the ability to excite a high fraction of electrons into the excited state. We have measured the temperature-dependent contribution to the linewidth $\gamma(T)$ at 189.6 GHz, with values of $\gamma(T)$ over 2 orders of magnitude smaller than previous direct measurements, into the ripplon scattering regime. We have made the first measurements of saturation and power broadening of the absorption line in this system, due to the finite occupancy of the first excited state. High levels of electron

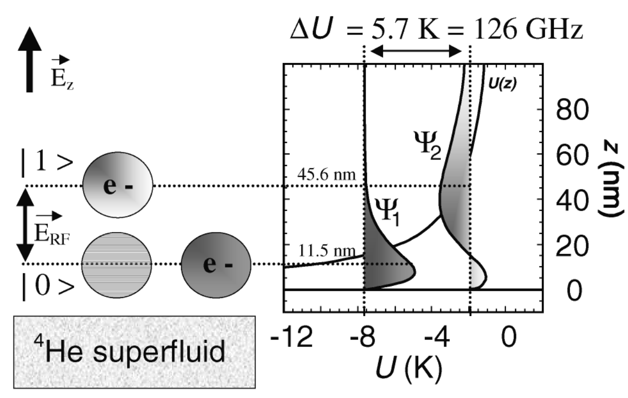

FIG. 1. The ground ($|0\rangle$, $m = 1$) and first ($|1\rangle$, $m = 2$) excited Rydberg states for electrons on helium for $E_z = 0$, showing the Coulomb potential $U(z)$ (solid line) and the electronic wave functions $\Psi_1(z)$ and $\Psi_2(z)$. The Stark effect comes from the different mean heights of the states above the helium (45.6–11.5 nm). Transitions from $|0\rangle$ to $|1\rangle$ are induced via a vertical microwave electric field E_{RF}.

excitation were achieved, very close to the limit for thermal equilibrium. The Rabi frequency was determined as a function of microwave power.

Power from a Gunn diode oscillator [12] was passed through a doubler (5 mW maximum output from 165 to 195 GHz) and transmitted down an overmoded waveguide, through thermal filters, into an experimental cell on a dilution refrigerator. The frequency of the Gunn oscillator $f = \omega/2\pi$ was phase locked to a 10 MHz quartz crystal resonator. Higher frequencies, up to 260 GHz, were obtained from a carcinotron source. The electrons were held above the liquid helium between capacitor plates 2 mm apart which formed a flat cylindrical cavity, 52 mm diameter. The microwaves were polarized vertically by a wire grid on the cavity input port and propagated horizontally through the cell to a low-temperature InSb Putley bolometer [13]. The vertical holding field E_z was swept by varying the potential difference V_z between the capacitor plates. Two techniques were used to measure the absorption linewidth. First, the holding field was sine-wave modulated (typically by 10 mV rms) at 5 kHz and the differential absorption signal measured by the Putley detector using a lock-in amplifier. This was integrated numerically to obtain the absorption line shape. In the second method, a larger amplitude (3 V) square wave pulse was applied, such that each half-cycle alternately sampled the resonant absorption or a region of very small absorption. The two methods gave identical line shapes.

The resonant frequency $f_{12} = \omega_{12}/2\pi$ versus E_z is shown in Fig. 2, in excellent agreement with previous experiments [2,3]. The integrated line shape above 1 K is close to the Lorentzian line shape

$$L(\delta) = \frac{\gamma/\pi}{\delta^2 + \gamma^2}, \tag{1}$$

where $\delta = \omega - \omega_{12}$. The half-width γ was measured in terms of the voltage V_z. The conversion factor to frequency is 2.44 GHz/V at 189.6 GHz (the slope of Fig. 2). Below 1 K, the linewidth decreases rapidly and becomes constant (typically 50 MHz). The line shape is no longer purely Lorentzian. Following Edel'man [4], we assume this is dominated by inhomogeneous broadening due to variations in the vertical holding field and, hence, the resonant frequency, across the electron sheet [14]. However, our experiments do not exclude a small temperature-independent contribution to the total intrinsic linewidth at low temperatures. The inhomogeneous absorption $\alpha_0(V_z)$ of the cell was measured at low temperatures, where the intrinsic linewidth is small. As the temperature increased, the experimental absorption line shape $\alpha(V_z, T)$ changed and broadened. The temperature-dependent contribution to the linewidth was obtained by convoluting a Lorentzian absorption, half-width $\gamma = \gamma(T)$, with the inhomogeneous low-temperature absorption, using γ as a best-fit parameter. The fit of the convoluted line shape to experiment was excellent at all temperatures, confirming that the temperature-dependent contribution is Lorentzian. The temperature-dependent contribution was less than the error bars below 300 mK, which was our reference temperature. Further details will be presented elsewhere.

By analyzing the line shape in this way, we measured the temperature-dependent contribution to the half-width $\gamma(T)$ as shown in Fig. 3. Above 1 K, scattering from ^{4}He vapor atoms dominates and is proportional to the vapor pressure, while below 1 K, the scattering is from surface

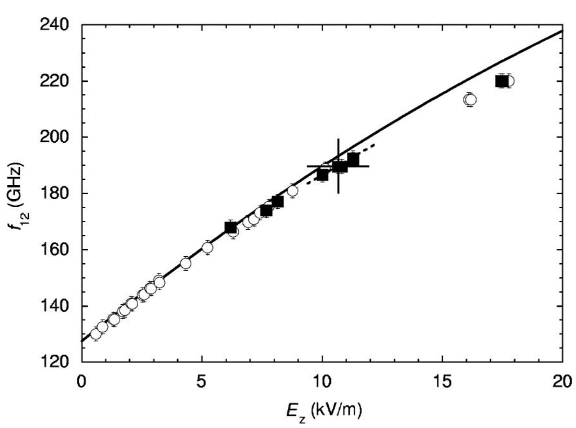

FIG. 2. Resonant frequency f_{12} versus E_z at 1 K, for low microwave powers (full squares). The open circles are from Grimes *et al.* [2] at 1.2 K. The solid line shows the theoretical calculations of [2]. The cross represents the frequency (189.6 GHz) we have chosen for our temperature/power characterizations. The dashed line is the slope of the experimentally measured curve at this point.

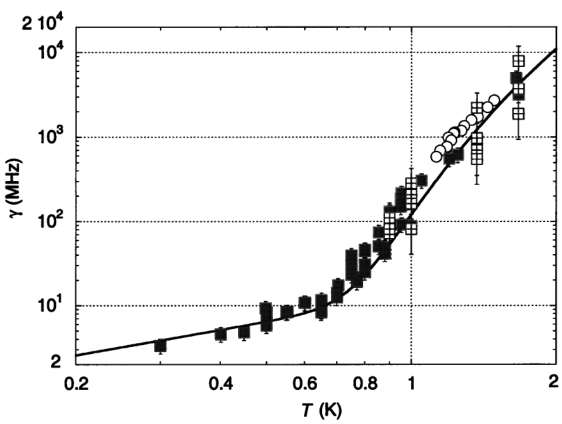

FIG. 3. Temperature-dependent contribution to the linewidth $\gamma(T)$ versus temperature for 189.6 GHz. Full squares are the low power measurements, obtained as explained in the text. The origin is determined by assuming that $\gamma(T) = AT$ below 700 mK. The circles are the data from [2]. The crossed squares are our results extracted from the power dependency (see text). The full line is Ando's theory [5].

waves (ripplons). The theory by Ando [5] gives a total intrinsic linewidth

$$\gamma(T) = AT + BN_{\text{gas}}, \tag{2}$$

where the first term is due to ripplon scattering and $N_{\text{gas}} \propto T^{3/2}\exp(-7.17/\text{T})$ is the number density of ^{4}He vapor atoms. The coefficients A and B depend on the holding field E_z. The experimental reference value of $\gamma(T)$ at 300 mK was determined by a linear fit of the temperature dependence below 700 mK, following Eq. (2).

Both inelastic and elastic collisions contribute to the linewidth. Inelastic collisions produce decay, or relaxation, of the excited state with a lifetime $\tau = 1/2\gamma_{\text{in}}$ [15] (the radiative lifetime is estimated to be very long, $\sim$0.1 s, in this system), while elastic collisions produce fluctuations in the energy levels and, hence, a linewidth γ_{el} (and also decoherence). The total Lorentzian half-width $\gamma = \gamma_{\text{in}} + \gamma_{\text{el}}$. The new experimental measurements cover three decades in $\gamma(T)$. They are in good agreement with previous data above 1.2 K and lie close to the theoretical values shown by the solid line in Fig. 3 [5].

Experimentally, the linewidth is independent of the microwave power, at low powers. As the power increases, the absorption line broadens and saturates. The saturation absorption $\alpha(P)$ at resonance is shown in Fig. 4, plotted versus the microwave input power, as measured by a power meter at the top of the cryostat. The transmission loss from the source to the detector was typically 20 to 30 dB. At low powers $\alpha = aP$ but it then saturates as $\alpha(P) = aP/(1 + aP/\alpha_{\max})$. Saturation is due to the finite occupancy of the excited state. The optical Bloch equations reduce to the rate equation [7] for the fractional occupancies of the ground and excited states, ρ_1 and $\rho_2 = 1 - \rho_1$

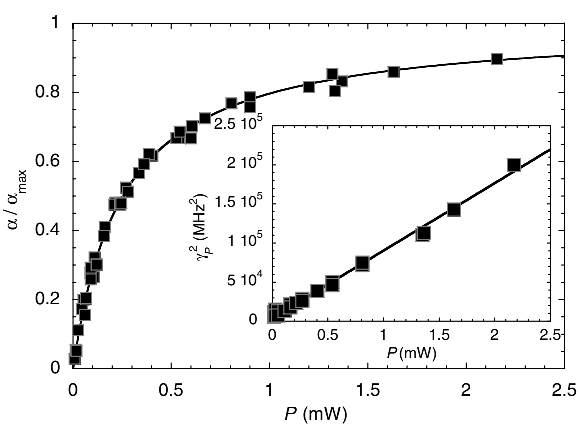

FIG. 4. Absorption saturation data $\alpha(P)/\alpha_{\max}$ at 0.9 K for 189.6 GHz, normalized to the maximum absorption. The input power P is measured at room temperature. The line is a fit explained in the text. Inset: Power broadening data γ_P^2 versus P, where the inhomogeneous broadening has been subtracted.

$$\frac{d\rho_2}{dt} = r(\rho_1 - \rho_2) - \frac{\rho_2}{\tau} = r(1 - 2\rho_2) - \frac{\rho_2}{\tau}, \tag{3}$$

where r is the rate for stimulated absorption and emission which is balanced by spontaneous emission. In dynamic equilibrium, $\rho_2 = r\tau/(1 + 2r\tau)$ with a saturation value of 0.5. This effect directly leads to power saturation and power broadening. The excitation rate can be expressed in terms of the Rabi frequency $\Omega = eE_{\text{RF}}z_{12}/\hbar$, where E_{RF} is the microwave field amplitude and z_{12} is the electric dipole length for the transition. The excitation rate $r = 0.5\Omega^2\gamma/(\delta^2 + \gamma^2)$ [7]. Hence, the power absorption $\alpha = N\rho_2/\tau$ is

$$\alpha = \frac{\frac{1}{2}N\gamma\Omega^2}{\delta^2 + \gamma^2 + \gamma\tau\Omega^2}, \tag{4}$$

where N is the number of electrons. This has the form shown experimentally in Fig. 4 with $\alpha_{\max} = N/2\tau = N\gamma_{\text{in}}$. The Lorentzian linewidth is now power dependent with $\gamma_P^2 = \gamma^2 + \gamma\tau\Omega^2 = \gamma^2 + bP$, where $P \propto E_{\text{RF}}^2 \propto \Omega^2$ is the incident microwave power. Power broadening is a direct measure of the Rabi frequency.

Figure 4 shows the experimental power dependent linewidth (with the inhomogeneous linewidth subtracted), plotted as γ_P^2 versus P, confirming the theoretical expression for power broadening. The intercept gives the low power value for $\gamma(T)$ as plotted in Fig. 3.

The fractional occupancy $\rho_2 = 0.5[1 - (\gamma/\gamma_P)^2]$ of the excited state can be obtained directly from the power broadening and has a maximum value of 0.489 for the data in Fig. 4, very close to the thermal equilibrium limit of 0.5. The Rabi frequency and the microwave electric field amplitude can also be obtained from the power broadening term $\Omega\sqrt{\gamma\tau}$ [15] which has a maximum value of 440 MHz for the data in Fig. 4. The dimensionless factor $\gamma\tau$ depends on the scattering and decay mechanisms producing the finite linewidth. Variational wave functions for the ground state $\Psi_1(z)$ and first excited state $\Psi_2(z)$ were calculated as a function of the electric holding field E_z [5]. Above 0.7 K, the linewidth is primarily due to electron scattering from ^{4}He vapor atoms which behave as point, or δ function, scattering centers. In this case, we estimate theoretically that $\gamma\tau = 7.1$ for $E_z = 0$, decreasing to 2.9 for $E_z = 1.06 \times 10^4$ V/m, corresponding to a resonant frequency of 190 GHz. This would give a maximum value for $\Omega = 260$ MHz for the data in Fig. 4. The dipole length for excitation $z_{12} = |\langle\Psi_1^*(z)|z|\Psi_2(z)\rangle|$ increases from 4.2 nm in zero pressing field to 5.1 nm at a resonant frequency of 190 GHz. The corresponding maximum microwave electric field amplitude would be $\simeq 200$ V/m. The power dependent data was also analyzed, using Eq. (4), to obtain values of $\gamma(T)$ above 0.9 K, as shown in Fig. 3, in good agreement with the low power data.

The total saturation power absorbed by the electrons is estimated as 70 nW at 0.9 K. No heating effects were apparent at this temperature, though these may become important at lower temperatures.

This analysis assumes that the response is independent of the electron density n. The strong electron-electron Coulomb interactions will not contribute to the linewidth directly (Kohn theorem) though they can mediate the scattering by other mechanisms, as in electronic transport in a magnetic field [16]. However, the electron-electron interactions will change the energies of the Rydberg states and, hence, the resonant frequency. Indeed, a basic idea for surface-state qubits is that the excitation of one electron will change the resonant frequency of its neighbor, via the Coulomb coupling [6]. For a typical electron density of $n = 0.18 \times 10^{12}$ m^{-2} and an interelectron distance of 2.5 μm, the Coulomb shift is about 12 MHz for nearest neighbors. This introduces a nonlinear term into Eq. (4) which becomes significant as $\gamma(T)$ becomes less than the Coulomb shift below 0.8 K [17]. Controlling these effects for interacting qubits is an overall objective. The power dependence in this region also changes, due to a change in the factor $\gamma\tau$ as the scattering mechanism changes.

Significant conclusions can be drawn concerning the use of these states as qubits for quantum processing. First, we have experimentally achieved the microwave field amplitudes and Rabi frequencies which were postulated by Platzman and Dykman [6] in their qubit proposal. The Rabi frequency would represent the clock frequency for qubits. Second, we have shown that the temperature-dependent contribution to the linewidth $\gamma(T)$ is indeed small at low temperatures, at least on bulk helium. However, a small temperature-independent contribution may also be present. Kirichek *et al.* [18] recently measured the scattering rate for surface-state electrons on both ^{3}He and ^{4}He from the linewidth of electron plasma resonances, down to 10 mK. On ^{4}He, they found excellent agreement with the theory of electron-ripplon scattering. For transport measurements, the scattering rate remains finite at the lowest temperatures. However, the microwave linewidth $\gamma(T)$ decreases linearly with temperature to low temperatures [5]. In our experiments at 100 mK, $f_{12}/\gamma \simeq 2 \times 10^5$ while $\Omega/\gamma \approx 300$. This demonstrates the high quality of the Rydberg states in this system. The small single ripplon-induced-decay would be further suppressed by localizing electrons in traps (as required for qubits) and by the application of a perpendicular magnetic field [6]. The present experiment gives grounds for optimism for the use of these states in electronic qubits, though much remains to be done.

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[1] For a collection of review articles, see *Two-Dimensional Electrons on Cryogenic Substrates*, edited by E. Andrei (Kluwer Academic Press, Dordrecht, 1997).
[2] C. C. Grimes, T. R. Brown, M. L. Burns, and C. L. Zipfel, Phys. Rev. B **13**, 140 (1976).
[3] D. K. Lambert and P. L. Richards, Phys. Rev. Lett. **44**, 1427 (1980); D. K. Lambert and P. L. Richards, Phys. Rev. B **13**, 3282 (1981).
[4] V. S. Edel'man, Zh. Eksp. Teor. Fiz. **77**, 673 (1979) [Sov. Phys. JETP **50**, 338 (1979)]; A. P. Volodin and V. S. Edel'man, Zh. Eksp. Teor. Fiz. **81**, 368 (1981) [Sov. Phys. JETP **54**, 198 (1981)].
[5] T. Ando, J. Phys. Soc. Jpn. **44**, 765 (1978).
[6] P. M. Platzman and M. I. Dykman, Science **284**, 1967 (1999); M. I. Dykman and P. M. Platzman, Fortschr. Phys. **48**, 1095 (2000); M. I. Dykman, P. M. Platzman, and P. Seddighrad (to be published).
[7] R. Loudon, *The Quantum Theory of Light* (Oxford Science, Oxford, 2000), 3rd ed.
[8] A. Abragam, *Principles of Nuclear Magnetism* (Oxford University, Oxford, 1985), reprinted.
[9] Lieven M. K. Vandersypen, Matthias Steffen, Gregory Breyta, Constantino S. Yannoni, Richard Cleve, and Isaac L. Chuang, Phys. Rev. Lett. **85**, 5452 (2000).
[10] Lieven M. K. Vandersypen, Matthias Steffen, Gregory Breyta, Constantino S. Yannoni, Mark H. Sherwood, and Isaac L. Chuang, Nature (London) **414**, 883 (2001).
[11] M. J. Lea, P. G. Frayne, and Yu. Mukharsky, Fortschr. Phys. **48**, 1109 (2000).
[12] Radiometer Physics Gmbh.
[13] QMC Instruments Ltd.
[14] The low-temperature broadening was minimum when the cell was leveled using segmented Corbino electrodes.
[15] Experimentally, we plot γ in frequency units (MHz). In the theoretical expression for $\gamma\tau$, γ is an angular frequency (rad/s).
[16] M. I. Dykman, C. Fang-Yen, and M. J. Lea, Phys. Rev. B **55**, 16 249 (1997).
[17] M. I. Dykman *et al.* (to be published).
[18] O. I. Kirichek, M. Saitoh, K. Kono, and F. I. B. Williams, Phys. Rev. Lett. **86**, 4064 (2001).