J Low Temp Phys (2010) 158: 324–330
DOI 10.1007/s10909-009-9956-1

Self-Sustained Microwave Absorption Induced by Extremely High Radiation Intensities in Surface Electrons on Liquid Helium

Denis Konstantinov · Kimitoshi Kono

Received: 15 June 2009 / Accepted: 1 September 2009 / Published online: 16 September 2009

Abstract A new nonlinear-optical absorption effect is observed in electrons bound to the liquid helium surface. We study absorption of mm-wave radiation due to resonant excitation of electron bound states. Below 1 K, almost all electrons occupy the ground state. Therefore, the system should be transparent for resonant radiation connecting any two excited states. On the contrary, we observe strong absorption peaks associated with transitions between the first excited and the higher excited states. We show that this anomaly results from the bistability of the electron system induced by extremely high radiation intensities and the long electron relaxation time.

Keywords 2D electrons on liquid helium · Microwave absorption · Optical bistability

PACS 67.90.+z · 73.20.-r · 78.70.Gq · 42.65.Pc

1 Introduction

Nonlinear-optical absorption effects, such as energy-transfer up-conversion and intrinsic optical bistability, are of great interest because of their applications in quantum counters, optical amplifiers, all-solid-state lasers etc. A particularly interesting effect occurs in some rare-earth-doped crystals where strong optical absorption is observed for laser pump light connecting only excited electronic states of rare-earth ions, while no transitions from the ground state are directly excited [1]. This counter-intuitive phenomenon is due to the so called photon avalanche, in which the cross-relaxation of ions self-sustains the excited state population. Intrinsic optical bistability has been theoretically predicted in this system [2] but remains to be observed.

D. Konstantinov (✉) · K. Kono
Low Temperature Physics Laboratory, RIKEN, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan
e-mail: konstantinov@riken.jp

In our experiment we study a very different system of strongly-correlated electrons bound to the free surface of liquid helium and pumped by the resonant millimeter microwaves (MW). Surprisingly, we observe a behavior reminiscent of that in rare-earth-doped materials. In particular, we observe anomalous and strong absorption of MW when the transition energy for two excited bound states of electrons is tuned in resonance with the radiation. We show that the extremely high radiation intensities used in our experiment and very long electron relaxation time provide a qualitatively new nonlinearity mechanism that leads to the occurrence of intrinsic optical bistability in this system.

Free electrons can be trapped at the vapor-liquid helium interface owing to the interplay between an attractive image potential and a very high repulsive barrier that prevent electrons from entering the liquid [3]. For the vertical motion, the energy spectrum of such surface electrons (SE) is a discrete Rydberg series, $\epsilon_n = -R/l^2$, where $l = 1, 2, \ldots$ is the quantum number and R is the effective Rydberg energy ($R/k_B \sim 7$ K for ^{4}He and 4 K for ^{3}He). The in-plane motion is free except for the scattering from He vapor atoms or surface capillary waves, ripplons. An exceptional purity of this system provides the unusually long electron relaxation time, reaching $\sim 10^{-7}$ s even at $T > 0.1$ K. The absorption of MW due to a resonant $l \to l'$ transition can be induced by tuning the transition energy $\epsilon_{l'} - \epsilon_l$ to match the radiation frequency. In the experiment, this is usually done by applying an electric field $E_\perp$ normal to the helium surface and shifting electron energy levels due to a linear Stark effect.

At low enough temperatures such that $T \ll R/k_B$, the thermal population of the excited ($l \geq 2$) states is negligible. Therefore, SE should not absorb radiation at frequencies matching transitions $l \to l'$ such as $2 \leq l < l'$. Nevertheless, below 1 K we observe strong absorption of MW that are in resonance with either $2 \to 3$ or $2 \to 4$ transition. A key phenomenon that allows us to explain this unexpected result is the resonant absorption-induced heating of electrons that was recently discovered in this system [4, 5]. It was shown that due to very slow electron energy relaxation, on the one hand, and a very efficient transfer of the excitation energy to the thermal energy of the in-plane motion, on the other hand, SE can be easily overheated causing significant population of the excited states. Below we present our experimental result and propose the theoretical explanation of the observed nonlinear effect.

2 Experimental

Our experimental method is based on the measurement of the absorption-induced change in SE conductivity. For the details of this method we refer to our previous publications [4, 6]. The experiment described in this report is done with SE held on the free surface of liquid ^{3}He placed approximately in the middle between two horizontal plates of a flat circular capacitor. A positive voltage is applied to the bottom plate to create an electric field $E_\perp$ normal to the surface. To excite a resonant $l \to l'$ transition between SE Rydberg levels, MW at frequency ω are passed between capacitor plates, and the transition frequency $\omega_{l'l} = (\epsilon_{l'} - \epsilon_l)/\hbar$ of SE is tuned in resonance with MW by varying the magnitude of $E_\perp$.

The resonance response is observed by measuring the low-frequency (100 kHz) in-plane magnetoconductivity σ_{xx} in a normal magnetic field B using a Corbino disk that constitutes the top plate of the capacitor. For a typical $B \sim 300$ G used in the experiment, the cyclotron frequency $\omega_c < \omega_p$, where the electron plasma frequency $\omega_p = (2\pi e^2 n_s^{3/2}/m)^{1/2}$ (m is the electron mass and n_s is the electron surface density.) Under such conditions, σ_{xx} of strongly-correlated SE follows the Drude low and is proportional to the single-electron scattering rate ν that electrons have in a zero magnetic field [7]. This rate, in turn, is determined by the electron temperature T_e [4, 6]. Thus the variation of σ_{xx} near the resonance reflects the degree of electron heating induced by resonant absorption and allows us to calculate T_e from the relative change of the scattering rate ν determined from the measured values of σ_{xx}.

3 Results

The typical experimental procedure consisted of measuring the conductivity signal using the Corbino disk while slowly increasing $E_\perp$ to drive electrons through resonance, while maintaining the radiation frequency and the radiation intensity fixed. The measurements done with MW at $\omega/2\pi = 105$ GHz are reported here. At this frequency, the maximum output of our source (W-band solid-state multiplier driven by RF/MW signal generator) is about 5 mW as given in the source specification.

For almost all radiation intensities, we observe only a single peak due to resonant $1 \to 2$ transitions. This is in complete agreement with our previous results [4, 6]. However, at the maximum radiation intensity a number of new features emerges. In particular, as $E_\perp$, and correspondingly the splitting between Rydberg levels, increases, we observe at least two more resonances identified with $2 \to 4$ and $2 \to 3$ transitions. An example of the recording at $T = 0.45$ K and at the electron density $n_s = 6 \times 10^6$ cm^{-2} is shown in Fig. 1. Here the Corbino response, which is proportional to the conductivity change $\Delta\sigma_{xx}/\sigma_{xx}$, is plotted as a function of $E_\perp$. The largest peak at $E_\perp \approx 45$ V/cm is due to the transition from the ground state to the first excited state, while an isolated peak at $E_\perp \approx 67$ V/cm matches in frequency the transition from the first to the third excited state. At much higher fields, we also observe a peak at $E_\perp \approx 188$ V/cm that matches the transition from the first to the second excited state. An example is shown in the inset of Fig. 1. We emphasize that at $E_\perp = 67$ V/cm and $E_\perp = 188$ V/cm the transition frequency $\omega_{21} \approx 114$ GHz and 170 GHz, therefore the transition from the ground state to the first excited state is completely detuned from the resonance with MW.

In addition, a number of side-peaks (one of such peaks appearing at $E_\perp \approx 39$ V/cm is shown in Fig. 1) were observed at very high radiation intensities. However, we believe that these features have no connection with the effect we report here, therefore we postpone the discussion of these side-peaks until our future publications.

Our measurements indicate that there is a certain threshold power above which $2 \to 4$ and $2 \to 3$ absorption peaks appear. Moreover, when approaching resonance from the low-field side, the absorption emerges suddenly, which is seen as an abrupt jump of the magnetoconductivity σ_{xx}. When sweeping through the resonance from the high-field side, we observe a similar jump on the opposite side of the resonance. Moreover, σ_{xx} does not follow the same dependence on $E_\perp$ upon reversing

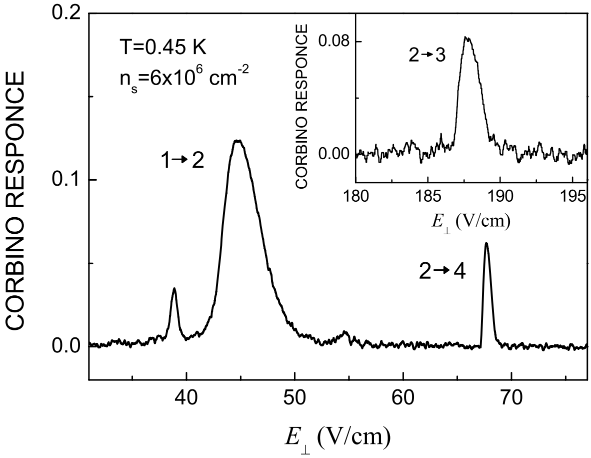

Fig. 1 Corbino response versus tuning field $E_\perp$ for SE on liquid ^{3}He. Measurements are done with MW at $\omega/2\pi = 105$ GHz at the maximum radiation intensity. The largest peak is due to resonant $1 \rightarrow 2$ transitions, while the peak on the far right is identified with resonant $2 \rightarrow 4$ transitions. In the inset: the resonant response recorded at higher $E_\perp$ and identified with $2 \rightarrow 3$ transitions. All measurements are done at $T = 0.45$ K with the electron density $n_s = 6 \times 10^6$ cm^{-2}

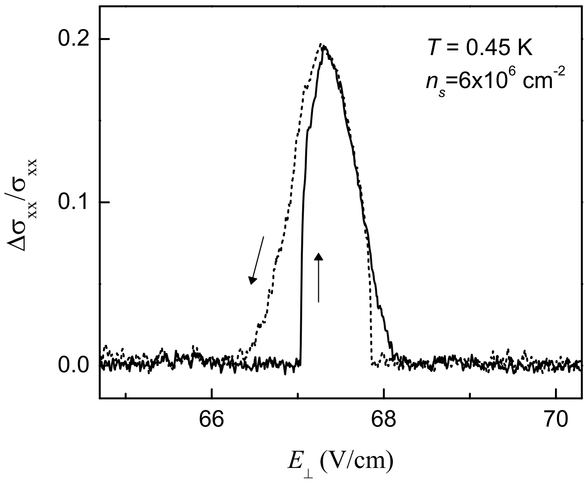

Fig. 2 Conductivity change $\Delta\sigma_{xx}/\sigma_{xx}$ due to radiation absorption for $2 \rightarrow 4$ transitions. Data are obtained in an upward sweep (*solid line*) and a downward sweep (*dashed line*) at the maximum radiation intensity and at $T = 0.45$ K. The electron density is $n_s = 6 \times 10^6$ cm^{-2}. The arrows indicate the direction of sweep. The temperature of the electron system at the absorption resonance is about 4 K as estimated from the value of $\Delta\sigma_{xx}/\sigma_{xx}$

the sweeping direction, thus demonstrating hysteresis. This anomalous behavior is illustrated in Fig. 2 where we show the conductivity change $\Delta\sigma_{xx}/\sigma_{xx}$ due to resonant $2 \rightarrow 4$ absorption measured at the maximum radiation intensity. We note that the sweeping rate is 10 times slower than that for the traces shown in Fig. 1. The response for $2 \rightarrow 3$ transitions (not shown) shows similar behavior.

4 Discussion

The straightforward estimation shows that at $T = 0.45$ K the fractional thermal occupancy of the first excited state is less than 10^{-5} and 10^{-7} for $E_\perp = 67$ V/cm and 188 V/cm, respectively. If we naively assume that electrons are always in equilibrium with the thermal bath, we should not observe any significant absorption of radiation tuned in resonance with transition between the first excited and higher-laying bound states. This is in acute contradiction with our experimental result.

However, as was recently shown [4, 5] the absorption of MW by SE is accompanied by the strong overheating of the electron system. The coupling between the vertical transitions and the in-plane motion of an electron provides an efficient transfer of the excitation energy to the energy of the in-plane motion. The fast exchange of the in-plane energy during electron-electron collisions allows to define the electron temperature T_e which is the same for all occupied electron states. Consequently, the electron scattering from vapor atoms or ripplons leads to thermal distribution over the ground and the excited states. It seems reasonable to suggest that besides a regime with $T_e \approx T$, there exists another stable regime with $T_e \gg T$, in which the population of the second excited state is high, the absorption due to transitions from this state to the higher-laying states is strong, and the overheating of the electron system is self-consistently large.

The existence of such a high-T_e state can be shown by considering the balance between the heating of SE by MW and cooling by the thermal bath. The energy balance equation can be written as [4, 5]

$$\hbar\omega r(\rho_{ll} - \rho_{l'l'}) = \nu_E k_B (T_e - T). \quad (1)$$

Here ρ_{ll} is the fractional occupancy of l-th state, ν_E is the electron energy relaxation rate, r is the rate of $l \to l'$ radiation-induced transitions, $r = 0.5\Omega^2\gamma/(\delta^2 + \gamma^2)$, where $\Omega = E_0 d_{ll'}/\hbar$ is the Rabi frequency (E_0 is the MW electric field and $d_{ll'}$ is the dipole transition matrix element), γ is the half-width of the absorption line, and $\delta = \omega - \omega_{l'l}$ is MW frequency detuning from the resonance. We note that the electron-electron interaction can cause the shift of the absorption line which depends on distribution over electron states [8]. However, the effect is sufficiently small for electron densities used in the present experiment, and we will ignore this shift in our calculations.

The stationary regime of the electron system can be found by solving (1) with respect to T_e. A way of obtaining the solution graphically is illustrated in Fig. 3 [5, 8]. The solution is defined by the intersection of a solid line and a broken line, which represent the cooling rate and the heating rate, respectively. These are calculated for SE on liquid ^{3}He at $T = 0.45$ K and for $E_\perp \approx 60$ V/cm. For the numerical evaluation, we assumed that ν_E and γ are determined by collisions with He vapor atoms. For the evaluation of r, we assumed that $\omega = \omega_{42}$, and used Ω as a parameter. Also, we assumed that ρ_{ll} is determined by the Boltzmann distribution. The heating rate for three different values of Ω are shown in Fig. 3. We find that there is a critical value of Ω above which there is a multiple solution of (1), as shown in Fig. 3 for $\Omega/2\pi = 15$ and 25 MHz. In particular, in addition to the solution $T_e \approx T$, there exist two more solutions with $T_e \gg T$. Such solutions are marked by circles in Fig. 3 for $\Omega/2\pi = 25$ MHz. The solution with higher T_e (solution S) corresponds to a stable regime of the electron system. The solution with lower T_e (solution U) corresponds to an unstable regime. For such a regime, any small deviations of T_e from the equilibrium value lead to the collapse of the system to one of the two stable regimes, as can be readily seen from Fig. 3. In particular, if T_e slightly increases, the heating rate becomes larger than the cooling rate, electrons heat up, and the system collapses

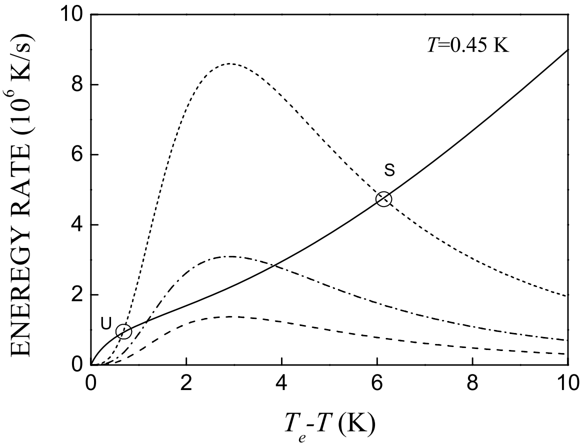

Fig. 3 Graphical solutions of the energy balance equation for $E_\perp \approx 60$ V/cm at $T = 0.45$ K. We assumed that MW are in resonance with $2 \to 4$ transitions. The solid line is the cooling rate $\nu_E k_B (T_e - T)$. The heating rate $\hbar\omega r(\rho_2 - \rho_4)$ is calculated for $\Omega/2\pi = 10$ MHz (*dashed line*), 15 MHz (*short dashed line*) and 25 MHz (*dash-dotted line*). At high Ω, besides the solution $T_e \approx T$, there exist two additional solutions, one of which is stable (solution S) and another is unstable (solution U). For $\Omega/2\pi = 25$ MHz, these are marked by *circles*

to the higher temperature stable state. Similarly, if T_e slightly decreases, the cooling rate becomes higher than the heating rate, and the system collapses to the lower temperature stable state.

The above solution of the energy balance equation suggests the occurrence of an intrinsic optical bistability, i.e. a co-existence of two stable regimes characterized by two different rates of radiation absorption, if the radiation intensity exceeds a certain threshold value. In one of these two regimes, SE are in thermal equilibrium with the thermal bath. Correspondingly, the thermal population of the first excited state is negligible, and the absorption of MW is vanishing. In another regime, $T_e \gg T$ and the first excited state is thermally populated. Correspondingly, the absorption of MW by electrons tuned in resonance for transitions from the first excited to the higher-laying states causes the heating and self-sustains high population of the first excited state.

The proposed theoretical model gives an excellent account for the behavior observed in the experiment. As discussed in Sect. 3, the strong anomalous absorption is seen only if MW power P exceeds a certain threshold. The critical value of Ω estimated from the threshold value of P is found to be in reasonable agreement with the calculations presented in this section. For the experimental data shown in Fig. 2, the temperature of the electron system is found to be about 4 K at the absorption resonance indicating the existence of the predicted stable high-T_e regime.

In summary, we have demonstrated the occurrence of the intrinsic optical bistability in surface electrons on liquid helium. The absorption of resonant MW is self-sustained by the strong radiation-induced overheating of the electron system and thermal redistribution of electrons over the surface bound states. The proposed theoretical model is in excellent agreement with the experimental result. We emphasize that the intrinsic optical bistability reported here is qualitatively different from the one discussed in our previous publication [8].

Acknowledgements We thank M. Dykman, M. J. Lea and Yu. P. Monarkha for valuable discussions. This work is partially supported by the Grant-in-Aids for Scientific Research from the MEXT.

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