Resonant Photovoltaic Effect in Surface-State Electrons on Liquid Helium

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(Dated: February 1, 2011)

We observed an electromotive-force effect induced by the resonant intersubband absorption of microwaves in surface-state electrons on liquid helium subjected to a perpendicular magnetic field. The effect emerges at the minima of radiation-induced conductance oscillations reported previously [D. Konstantinov and K. Kono, Phys. Rev. Lett. **105**, 226801 (2010)] and is characterized by a nonequilibrium spatial distribution of electrons in the confining electrostatic potential.

PACS numbers: 73.20.At, 73.21.-b, 72.20.My, 78.70.Gq

Under electromagnetic irradiation, a net flow of charged particles may occur even in the absence of an average macroscopic force acting on them. As a result, the separation of positive and negative carriers causes an electrical potential difference, known as an electromotive force (EMF), to develop across the system. In semiconductors, such a photovoltaic effect can result, for example, from the diffusion of nonequilibrium charged carries produced by light or their separation at p-n junction [1]. The latter mechanism makes it possible to convert the energy of radiation into electricity and has important applications in photodiodes and photovoltaic cells. Recently, significant effort has been made to increase the efficiency of solar cells by employing new concepts based on, for example, nanostructured materials [2], hot-carrier transfer [3], and surface plasmon resonance [4]. In a number of experiments [5–8], photovoltaic effects were reported in conjunction with microwave-induced oscillations and zero-resistance states (ZRS) in a two-dimensional electron system (2DES) in high-mobility GaAs/AlGaAs heterostructures [9, 10].

In this Letter, we report the first observation of an EMF effect induced in surface-state electrons (SSE) on liquid helium by the interaction of electrons with a resonant microwave radiation field. In such a system, surface state subbands with energies ϵ_n (n = 1, 2, ..) are formed owing to the attractive image force, the repulsive surface barrier, and an electric field E_{\perp} applied perpendicular to the surface [11]. Below 1 K, almost all electrons are frozen into the lowest subband. Recent work has provided evidence that an effect similar to the microwaveinduced ZRS of a quantum degenerate 2DES occurs in a nondegenerate system of SSE on helium [12]. In the experiment in Ref. 12, the intersubband $n = 1 \rightarrow 2$ transition was excited using microwaves with angular frequency ω such that $\hbar \omega = \epsilon_2 - \epsilon_1$. Under such conditions, the longitudinal conductivity σ_{xx} , measured using capacitively coupled Corbino electrodes, underwent oscillations upon varying the perpendicular magnetic field B, exhibiting a sequence of minima shifted to lower values with respect to B satisfying the relation $\omega/\omega_c = l$, where $\omega_c = eB/m$ is the cyclotron frequency and $l = 4, 5, \dots$ The vanishing

conductivity $\sigma_{xx} \to 0$ at the minima implied vanishing resistivity ρ_{xx} in accordance with the standard tensor relation. Remarkably, the vanishing conductivity exhibited a hysteretic response, indicating the intrinsic instability of the electronic states. Here, we demonstrate that at the minima of the conductivity oscillations the radiation causes the separation of electrons from the neutralizing background provided by the positively charged metallic electrodes, resulting in a large EMF on the order 1 V across the electron layer. Our finding suggests a novel photovoltaic effect occurring in a 2DES subjected to a perpendicular magnetic field.

Figure 1 shows a schematic diagram of the experimental apparatus. Liquid ³He is condensed into a cell (a flat cylindrical cavity), which contains a sintered silver heat exchanger to ensure sufficient heat contact between the liquid and the cell body. The liquid level is set midway between two parallel plates, each having a diameter of 26 mm, separated by d = 2.6 mm. The bottom plate is divided into electrodes B and G_1 by a gap at a distance of 10 mm from the center of the plate, while the top plate is divided into electrodes C_1 , C_2 and G_2 by two gaps at distances of 7 and 10 mm from the center. Each of the three gaps has a width of 0.2 mm. Electrodes C_1 and C_2 form a Corbino disk (dc-grounded), from which σ_{xx} for the SSE can be determined as described previously [12]. Electrons are produced by briefly heating a tungsten filament F, while a positive bias $V_{\rm B} = 0.4$ V is applied to B with all other electrodes grounded. The amount of charge, which forms a circular pool on the liquid surface above electrode B, is determined by assuming the condition of complete screening of the electric field above the liquid and corresponds to about 5×10^6 electrons.

The positively biased electrodes provide a neutralizing background, which counteracts the Coulomb repulsion between SSE and prevents them to spread away. Below 1 K, the average thermal kinetic energy of an electron moving along the surface does not exceed 10^{-4} eV, which is much less than the typical difference of its electrical potential energy across the layer. Therefore, the effects associated with the diffusion of electrons are neglected here. In the absence of radiation, the equilibrium

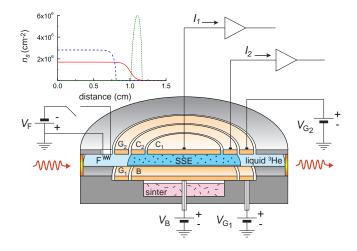


FIG. 1: (color online). Schematic diagram of the experiment. SSE are accumulated on the surface of liquid ³He and are excited by microwaves (wavy arrows), which enter and leave the cell through a pair of sealed windows. In the inset: areal density of SSE versus distance from center obtained numerically for $V_{\rm B} = 0.4$ V, $V_{\rm G_1} = V_{\rm G_2} = 0$ (solid line, red); $V_{\rm B} = 6.4$ V, $V_{\rm G_1} = V_{\rm G_2} = 0$ (ashed line, blue); and $V_{\rm B} = V_{\rm G_1} = 6.4$ V, $V_{\rm G_2} = 3.0$ V (dotted line, green).

spatial distribution of electrons in the confining electrostatic potential can be obtained numerically using the Green function method [13]. Examples of the equilibrium density profile calculated for 5×10^6 electrons and for different electrode biases are shown in the inset of Fig. 1.

Electrons are tuned for the intersubband $n = 1 \rightarrow 2$ resonance with the applied microwaves by adjusting the bias $V_{\rm B}$, which changes the electric field E_{\perp} and shifts the subband energies because of the Stark effect [14]. For $\omega/2\pi = 90.9$ GHz employed in this experiment, the resonance is centered at $V_{\rm B} = 6.4$ V. In the case of 5×10^6 electrons and all electrodes grounded except for electrode B, this bias corresponds to the areal density of electrons $n_s = 2.8 \times 10^6 \text{ cm}^{-2}$ (see the inset of Fig. 1). Previously [12], a photoinduced change in σ_{xx} was observed upon CW irradiation and in the presence of a driving electric field. The present work aims at detecting the photoinduced motion of the electrons in the absence of such a field. This is stimulated by the recent theoretical proposal that ZRS of 2DES in heterostructures could be explained by the stabilization of edge trajectories under microwave irradiation [15]. Indeed, such a mechanism might lead to an accumulation of charge at the edges and thus to an EMF. In this experiment, the incident microwave power is modulated using a low-frequency (0.5-2 Hz) square waveform, and the current signals I_1 and I_2 induced in electrodes C_1 and C_2 , respectively, are recorded using a digital storage oscilloscope following the current preamplifiers (100 Hz bandwidth). Alternatively, the current ΔI between electrodes C_1 and C_2 is measured

using a conventional lock-in amplifier.

Figure 2(a) shows an example of the transient current signals I_1 and I_2 obtained at T = 0.2 K, $n_s =$ 2.8×10^6 cm⁻², and B = 0.62 T. The value of B corresponds to the l = 5 conductance minimum. For reference, the modulation of the microwave power is shown in Fig. 2(b). Each trace represents the average over about 500 sweeps. The signals are comparable in magnitude and have opposite signs. From this result, we obtain the following qualitative picture. Upon switching the power on, the SSE are forced to flow along the surface, causing the depletion (accumulation) of the charge in the central (peripheral) region of the electron pool. Correspondingly, the positive (negative) current is induced in electrode C_1 (C_2) by the flow of the image charge. The surface charge flows until a new spatial distribution of SSE in the unchanged confining electrostatic potential is established, after which the currents I_1 and I_2 becomes zero. Correspondingly, the displacement of electrons with respect to the neutralizing background induces a nonzero electric field, and therefore EMF, to develop in the charged layer. Upon switching the power off, the displaced surface charge flows back to restore the equilibrium distribution of SSE, and a negative (positive) current consisting of the image charge is induced in C_1 (C_2).

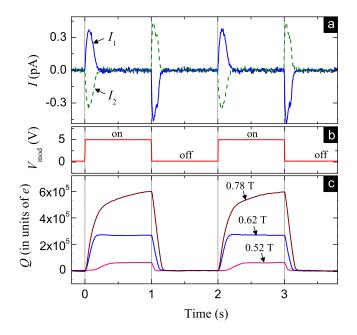


FIG. 2: (color online) (a) Transient signals I_1 (solid line) and I_2 (dashed line) induced in electrodes C_1 and C_2 , respectively, by the photoinduced flow of the surface charge at $n_s = 2.8 \times 10^6$ cm⁻², T = 0.2 K, and B = 0.62 T. (b) Square wave used to control the operation of the microwave source. The latter is switched on (off) at a high (low) signal level. (c) Cumulative charge Q obtained by integrating the current I_1 at three values of B corresponding to l = 4 (brown line), l = 5 (blue line), and l = 6 (pink line) conductance minima.

An important quantity, which determines the magnitude of the EMF, is the fraction of the surface charge displaced under irradiation. The latter can be estimated from the cumulative charge Q flowing, for example, through electrode C_1 . Q (in units of the elementary charge e > 0 is shown in Fig. 2(c) for three values of B corresponding to the conductance minima l = 4, 5, and 6. The total image charge induced on electrode C_1 in the dark can be determined from the following procedure. At $V_{\rm B} = V_{\rm G_1} = 6.4$ V, we apply a positive bias of 3.0 V to the electrode G_2 . As a result, the surface charge is pulled away from the center to form a ring beneath electrode G₂, as confirmed numerically by calculating the charge profile (dotted line in the inset of Fig. 1). When the sign of V_{G_2} is reversed, the charge is pushed back into the region beneath electrode C_1 , inducing a current in the electrode. Integrating this current, we find that a total image charge of $1.0 \times 10^6 e$ is induced in electrode C₁ by the SSE. This value is in good agreement with an estimate from a simple model of a charged particle between two conducting plates, as well as precise calculations using the Green function method. The comparison of this value with Q in Fig. 2(c) shows that a significant fraction (more than 50%) of the surface charge beneath electrode C_1 can be displaced upon irradiation. For our density, a 50% displacement yields an EMF of about 0.4 V, which corresponds to the difference in the potential energy of an electron markedly exceeding other characteristic energies such as, for example, the photon energy $\hbar\omega \approx 0.38$ meV.

Figure 3 shows a comparison between σ_{xx} measured under CW irradiation and ΔI recorded under 100% modulation of the incident microwave power. Both sets of data are obtained upon increasing the magnetic field Bat T = 0.2 K and at the same level of microwave power. A nonzero signal ΔI is observed only in the intervals of B near the conductance minima corresponding to l = 4, 5, 6, and 7. As described above, this signal is due to the currents induced in electrodes C_1 and C_2 by the displacement of the surface charge. No signal is observed when electrons are tuned away from the intersubband resonance using the microwaves by changing the electrical bias $V_{\rm B}$. Simultaneously, the detuning results in the complete disappearance of the magnetoosillations and zeroconductance states [12]. Importantly, the disappearance of ΔI in the SSE tuned away from the resonance, as well as when the surface is completely discharged by reversing the sign of $V_{\rm B}$, confirms that the current is not produced in the conducting electrodes by, for example, microwaveinduced heating, vibrations, or other effects.

The inset of Fig. 3(b) shows the signal ΔI in a narrow range of B near the l = 5 conductance minimum. The signal emerges abruptly upon slowly increasing B. This feature is consistent with the abrupt drop of σ_{xx} to zero recorded upon the upward sweep of B (solid circles). Upon the downward sweep of B, σ_{xx} (open circles) exhibits hysteresis, which may be due to the bistability of

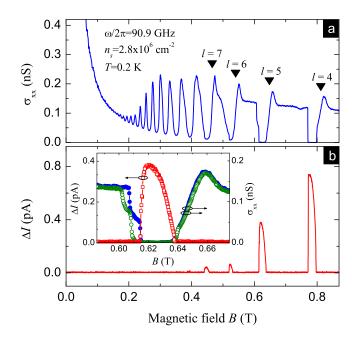


FIG. 3: (color online) (a) Microwave-induced magnetooscillations of SSE obtained for $n_s = 2.8 \times 10^6$ cm⁻² at T = 0.2 K under CW irradiation. (b) Current signal ΔI obtained under 100% modulation of the microwaves, and for the same n_s and T. Black triangles indicate the values of B such that $\omega/\omega_c = l$. In the inset: ΔI (left axis) in the range of B. For comparison, σ_{xx} (right axis) is shown for upward (solid circles) and downward (open circles) sweeps of B.

the electronic states [12]. In contrast, the variation of ΔI with *B* does not show any dependence on the direction of sweeping. The absence of hysteresis of ΔI might result from the modulation of the microwave power, which is repeatedly switched on and off, thus making the observation of the metastable regime impossible.

The kinetics of the radiation-induced displacement of surface charge strongly depends on different parameters such as the microwave power, the density of electrons, and the index l. The cumulative charge measured for $n_s = 2.8 \times 10^6 \text{ cm}^{-2}, B = 0.78 \text{ T}, \text{ and at four different}$ power levels is shown in Figure 4. For 2DES in Fig. 1, the typical time required to recover the equilibrium spatial distribution, the relaxation time $\tau_{\rm d}$, can be estimated as C/σ_{xx} , where C is the capacitance between the charged layer and the surrounding electrodes. For $C\sim 1~{\rm pF}$ and $\sigma_{xx} \sim 10^{-10}$ S, this gives $\tau_{\rm d} \sim 10^{-2}$ s, which is in agreement with the fall time of Q upon switching the power off, as shown in Fig. 4. In contrast, the time required to establish the redistribution of electrons across the laver upon switching the power on, as can be seen from the rising edge of Q, can exceed 1 s at sufficiently low powers. Even at high powers, the build-up of EMF is slowed by either increasing n_s or choosing the conductance minima of a high index l. Remarkably, a time delay of up to 0.1 s between the application of microwaves and the onset of

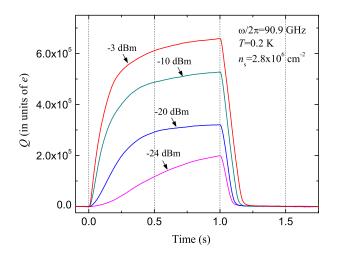


FIG. 4: (color online) Cumulative charge in electrode C_1 at B = 0.78 T for four power levels indicated for each curve. The power is switched on (off) at t=0 (1.0 s).

the charge motion can be observed, as shown, for example, by the data labeled as 0.52 T in Fig. 2(c).

In summary, the photoresponse of SSE on liquid helium in the absence of a dc electrical driving reveals the existence of a microwave-induced current in the electron layer. The displacement of charge produces EMF, which corresponds to the difference in the electron potential energy on the order 1 eV, which exceeds, for example, its thermal kinetic energy, cyclotron energy, or intersubband energy by several orders of magnitude.

The origin of such a strong effect is presently unclear. Radiation can exert an optical force on a charged particle, as is widely used for the cooling and trapping of atoms and ions [16]. This force can be estimated as the expectation value $-\langle \partial H / \partial x \rangle$, where H describes the interaction of the particle with the radiation field and xis the distance along the surface. Our estimations show that the optical force is too weak to directly account for the magnitude of the EMF reported here. The appearance of this effect at the minima of the conductance oscillations indicates the inherent relationship between EMF and σ_{xx} . In addition, an important question is the relation of the effect reported here to photovoltaic effects. which accompany the microwave-induced resistance oscillations in GaAs/AlGaAs heterostructures [5–8]. In Ref. 7, the oscillations of photocurrent and photovoltage were interpreted in terms of built-in electric fields due to band bending at contacts. Clearly, such fields do not exist in our system, and here we omit such a mechanism from further consideration. The microwave field gradient in the near-contact region can generate a ponderomotive force acting on 2DES in heterostructures, which may be a possible explanation for the ZRS in this system [17]. Such an effect is unlikely to occur in our system because the SSE are not in contact with the conducting electrodes. Recently, it was proposed that

microwave-induced oscillations of σ_{xx} in SSE on helium originate from the intersubband scattering of electrons under the condition of nonequilibrium filling of the excited subband [18]. This mechanism can result in an absolute negative σ_{xx} , causing current instability and the formation of an inhomogeneous pattern of current domains [19]. Such an effect is believed to be responsible for the EMF responses observed in Refs. 5 and 8, and may be a possible explanation for the redistribution of charge reported here. The microwave stabilization of edge trajectories, which was proposed recently to explain ZRS in heterostructures, naturally gives the correct direction of the charge displacement [15]. The creation of ballistic channels at the sample edges due to microwave irradiation combined with a pumping mechanism through the inter-subband excitation might lead to an accumulation of charge at the edges. All the possible outlined scenarios require thorough theoretical investigation.

We thank H. Bouchiat and D.L. Shepelyansky for helpful discussions. This work was supported in part by Grant-in-Aids for Scientific Research from MEXT. D.K. acknowledges an Incentive Research Grant from RIKEN.

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