Aloof electron probing of in-plane SPV charge distributions on GaAs surfaces

Zilin Chen,1,2 Wayne Cheng-Wei Huang,3,2† and Herman Batelaan2‡

1Department of Physics and Astronomy, Northwestern University, Evanston, Illinois 60208, USA
2Department of Physics and Astronomy, University of Nebraska-Lincoln, Lincoln, Nebraska 68588, USA
3Department of Physics, National Tsing Hua University, Hsinchu 30013, Taiwan (R.O.C.)

The motion of free electrons moving parallel and above a semiconductor surface can be influenced by shining laser light onto the surface. Here we report strong deflection of aloof electrons by an undoped GaAs surface illuminated with a 633 nm laser. The deflecting electric field from the surface photovoltaic charges extends 100 µm into the vacuum. As surface photovoltage (SPV) is sensitive to the electronic states of the GaAs surface, the aloof electron beam serves as a probe for SPV charge dynamics at the mesoscopic length scale. The observed in-plane SPV charge distribution persists beyond 1 second after the laser beam is blocked. Our work suggests the possibility of writing designed 2D charge patterns on semiconductor surfaces with a scanning laser beam, providing unusual flexibility for electron beam manipulation.

I. INTRODUCTION

The surface photovoltage (SPV) can facilitate electrostatic near fields above optically illuminated semiconductor surfaces. The near fields are important for SPV spectroscopy [1], for understanding charge separation and recombination processes in solar cells [2], for electron beam manipulation [3], and for studying quantum decoherence and dissipation of free electrons [4, 5]. In this study, we probe the SPV near fields on an undoped single-crystalline GaAs (110) surface through the deflection of aloof electrons. We observe strong deflection when the surface is under superband or subband photoexcitation. As SPV near fields depend on the transport properties and electronic states of the surface, our approach serves as a probe for SPV charge dynamics.

Recent developments on SPV measurement include SPV microscopy [2] and scanning ultrafast electron microscopy [6]. These methods are direct probes of the 2D photovoltaic charge distribution at the microscopic scale. Our approach complements the existing methods in that we measure the 3D SPV near fields at the mesoscopic length scale. Specifically, the vertical electron deflection due to SPV near fields is proportional to the 1D integral of the SPV charge distribution along the direction of electron beam propagation [7]. The vertical deflection we report here is up to 200 µm (7 beam diameters) at 40 cm from the GaAs surface. It can thus be used with an aperture to switch an electron beam on and off. Also, the SPV near field extends 100 µm into the vacuum, providing a long working distance. Our method is robust, requires no nanofabrication and works at modest vacuum (10⁻⁶ Torr).

The manipulation of free electrons by laser-illuminated material structures is of general interest and many examples exist for such technology. For example, the electron beam, and many copies thereof, can be steered by laser light in the presence of a surface and used for multibeam electron lithography [8]. In addition, control of electron motional states in dielectric laser accelerators [8], control of attosecond electron dynamics near a nanotip [9, 10], and laser-induced phase modulation of an electron wave [11] are but a few examples of exquisite motion control through photoinduced near fields. As SPV charge distributions is closely linked to the intensity profile of the laser light, simulation can be used to guide the development of a designed near field structure, providing another useful tool for optical control of free electrons.

A detailed understanding of photoinduced near fields and the ensuing change of surface resistivity are also needed for attaining controlled electron-surface decoherence [12-15]. This approach was first proposed by Zurek [4], intended for testing Caldeira and Leggett’s quantum dissipation theory [5]. In our experiment, we found that the electron diffraction pattern can be strongly distorted by the gradient force of the SPV near field, but the beam coherence is not affected by the light-modulated surface resistivity.

II. RESULTS AND DISCUSSIONS

A. Superband Photoexcitation

In our experiment (Fig. 1), in-plane SPV charge distributions are probed with a diffracted electron beam through vertical beam displacement. We use rate equations to model the photovoltaic carrier dynamics [11-14] and an electron trajectory simulation [15] to compute the near-field interaction that leads to the beam displacement.

As the undoped GaAs has a bandgap of 1.42 eV (873 nm), illuminating the surface with a 633 nm continuous-wave He-Ne laser creates free electrons and holes through superband excitation. The generated electrons and holes are close to the surface because the laser penetration depth in this case is only 300 nm [17]. This results in most free electrons being trapped in the surface states, while the free holes diffuse to the surrounding areas (Fig. 2(a)). The direction of electron beam displace-
FIG. 1. Schematic of the electron deflection experiment. A thermionic electron gun (top left) emits electrons at an energy of 1.67 keV. Two collimation slits separated by 250 mm limit the transverse momentum spread of the beam and deliver an electron beam with a transverse spatial coherence of ∼ 500 nm to a nanofabricated grating. The grating has a periodicity of 100 nm and transmits 50% of the electron beam. The diffracted electron beam passes parallel to and over a 10 mm long undoped GaAs surface. The surface is illuminated with an elliptically shaped laser beam. The laser beam widths parallel and transverse to the direction of electron beam propagation are 5 mm and 100 µm, respectively. The laser wavelength is 633 nm for superband excitation or 1064 nm for subband excitation. The nominal laser power is 1 mW. The laser produces a SPV charge distribution (purple and blue shades) whose electrostatic near field deflects the free electrons at an electron-surface distance up to 100 µm. The deflected electron beam is magnified with a quadrupole lens and recorded with an imaging detector (bottom right).

ment (Fig. 2(b)) indicates that trapped electrons locate at the center of the charge distribution, while the side wings are populated with free holes. In the equilibrium state, with the laser continuously on, the internal electric field within the charge distribution becomes strong enough to keep the holes from diffusing further away. The magnitude of electron deflection saturates at low laser power (1 mW) and is independent of laser polarization.

We use rate equations [1, 16, 18] to obtain the equilibrium density distributions for free electron $n(x)$, free hole $p(x)$, and trapped electron $n_t(x)$,

$$\frac{dn(x)}{dt} = \frac{1}{e} \frac{dJ_n(x)}{dx} + G_n(x) - R_n(x), \quad (1)$$

$$\frac{dp(x)}{dt} = -\frac{1}{e} \frac{dJ_p(x)}{dx} + G_p(x) - R_p(x), \quad (2)$$

$$\frac{dn_t(x)}{dt} = G_t(x) - R_t(x), \quad (3)$$

where $G_i(x)$ and $R_i(x)$ denote the generation and recombination rates, respectively, and $e$ is the electron charge unit. The charge current density $J_i(x)$ depends on the balance between diffusion and the drift caused by the internal electric field $E(x)$,

$$J_n(x) = e\mu_n n(x) E(x) + e D_n \frac{dn(x)}{dx}, \quad (4)$$

$$J_p(x) = e\mu_p p(x) E(x) - e D_p \frac{dp(x)}{dx}, \quad (5)$$

where $\mu_n = 8000$ cm$^2$/Vs and $\mu_p = 300$ cm$^2$/Vs are the mobilities of free electrons and holes in undoped GaAs. The corresponding diffusion coefficients are $D_n = 200$ cm$^2$/s and $D_p = 10$ cm$^2$/s. The internal electric field $E(x)$ is determined by the total charge density $\rho(x) = e (p(x) - n(x) - n_t(x))$. The thermal recombination rate $R_n(x) = r_{n\to p}^{th} + r_{n\to t}^{th}$ in Eq. (1) is the sum of the thermal transition from the conduction band to the valence band $r_{n\to p}^{th} = \tau_{n\to p}^{th} n(x)p(x)$ and the thermal
capture rate from the conduction band to the surface trapping states $r_{n ightarrow t}^{th} = c_{n ightarrow t}^{th} n(x) \left( N_t - n_t(x) \right)$, where $N_t$ is the trapping state density \[1\]. On the other hand, the generation rate $G_n(x) = g_{p ightarrow n}^{th} + g_{p ightarrow n}^{opt} + g_{t ightarrow n}^{th} + g_{t ightarrow n}^{opt}$ has four contributions. The $g_{p ightarrow n}^{th}$ and $g_{p ightarrow n}^{opt}$ terms correspond to thermal and optical excitation from the valence band to the conduction band. Similarly, the $g_{t ightarrow n}^{th}$ and $g_{t ightarrow n}^{opt}$ terms characterize thermal and optical excitation rates from the trapping states to the conduction band. These four terms are given by

$$g_{p ightarrow n}^{th} = c_{n ightarrow p} n_0 p_0,$$

$$g_{p ightarrow n}^{opt} = F(1 - R) \eta e^{-x^2/\omega_z^2},$$

$$g_{t ightarrow n}^{th} = \sigma_{t ightarrow n} n_t(x),$$

$$g_{t ightarrow n}^{opt} = F \sigma_{t ightarrow n}^{opt} n_t(x).$$

where $n_0$ and $p_0$ are the intrinsic carrier densities ($\sim 10^6 \text{cm}^{-3}$) in the absence of laser illumination, $F$ is the laser photon flux ($\sim 10^{18} \text{cm}^{-2} \text{s}^{-1}$), $R = 0.3$ is the GaAs reflectivity at 633 nm \[19\], $\eta = 0.7$ is the quantum efficiency \[20\], $\omega_z = 100 \mu \text{m}$ is the laser beam width transverse to the direction of electron beam propagation, $\sigma_{t ightarrow n}^{th}$ and $\sigma_{t ightarrow n}^{opt}$ are thermal and optical excitation rates from the trapping states to the conduction band. In Eqs. \[2\] and \[3\], the corresponding generation and recombination rates are

$$G_n(x) = g_{p ightarrow n}^{th} + g_{p ightarrow n}^{opt} + g_{t ightarrow n}^{th} + g_{t ightarrow n}^{opt}.$$
$G_p(x) = g_{p\rightarrow n}^{th} + g_{p\rightarrow n}^{opt}$, \hspace{1cm} (7a)

$R_p(x) = r_{n\rightarrow p}^{th}$, \hspace{1cm} (7b)

$G_t(x) = r_{n\rightarrow t}^{th}$, \hspace{1cm} (7c)

$R_t(x) = g_{t\rightarrow n}^{th} + g_{t\rightarrow n}^{opt}$ \hspace{1cm} (7d)

We obtain the total charge density $\rho(x)$ by solving the coupled rate equations Eqs. (1)-(3) with parameters given above. To compare the SPV charge distribution $\rho(x)$ with the electron deflection data, we construct an electron trajectory simulation \[15\] using an electrostatic field derived from the charge distribution. In Fig. 3(b), we find a good agreement between our model and the deflection data.

The superband model also predicts slow relaxation of the SPV charge distribution after the laser beam is blocked. While the SPV charge distribution is usually established in microseconds or faster \[6\], \[7\], its relaxation can take seconds due to surface trapping states \[16\], \[21\]. After blocking the laser with a mechanical chopper, we observe that the electron deflection persists for 1 second. This opens an interesting prospect that scanning a laser beam at a rate above 10 Hz can result in a “programmable” 2D surface charge pattern. The resulting SPV near field and its gradient may be exploited for building deformable electron-optical elements such as electrostatic lenses or deflectors.

### B. Subband photoexcitation

In order to verify the relation between slow relaxation and surface trapping states, we illuminate the GaAs surface with a 1064 nm continuous-wave laser. Even though the 1064 nm photon energy is less than the GaAs bandgap, free electrons and holes can still be generated via EL2 defect-assisted subband photoexcitation \[22\], \[23\]. As the laser penetration depth at 1064 nm exceeds the thickness of our GaAs sample (500 µm), the majority of free electrons and holes are generated in the bulk, and surface trapping states can be ignored. This allows us to measure the SPV relaxation time in the absence of surface trapping states. The long penetration depth of 1064 nm laser is confirmed by observing a red diffuse spot at an electron-surface distance up to 100 µm. The electrostatic near field of SPV charges causes vertical deflection of a diffracted electron beam at an electron-surface distance up to 100 µm. The presence of surface trapping states significantly impedes the electron-hole recombination at the surface, making the lifetime of the in-plane SPV charge distribution exceed over 1 second. For 1064 nm subband excitation, where no surface trapping states are involved, the SPV relaxation time is measured to be shorter than 0.6 ms. It is perhaps interesting to contemplate scanning a laser beam so that a designed 2D charge pattern could be “written” on the undoped GaAs surface via superband excitation. The gradient of the SPV near field can act as an electrostatic lens for the electron beam passing over the surface. Such “programmable” electron-optical elements may add an interesting approach to optical control of free electrons.

### III. CONCLUSION

In summary, we perform an electron deflection experiment with an undoped GaAs (110) surface. The surface is optically illuminated by low-power lasers for either superband or subband photoexcitation. We find good quantitative agreement between our experimental results and existing photovoltaic models. For 633 nm superband excitation, we observe a narrow central region populated with trapped electrons and equally narrow side wings of free holes. The electrostatic near field of SPV charges causes vertical deflection of a diffracted electron beam at an electron-surface distance up to 100 µm. The presence of surface trapping states significantly impedes the electron-hole recombination at the surface, making the lifetime of the in-plane SPV charge distribution exceed over 1 second. For 1064 nm subband excitation, where no surface trapping states are involved, the SPV relaxation time is measured to be shorter than 0.6 ms. It is perhaps interesting to contemplate scanning a laser beam so that a designed 2D charge pattern could be “written” on the undoped GaAs surface via superband excitation. The gradient of the SPV near field can act as an electrostatic lens for the electron beam passing over the surface. Such “programmable” electron-optical elements may add an interesting approach to optical control of free electrons.
ACKNOWLEDGMENTS

We thank Prof. H. Ruda for advice on the effect of EL2 states. The funding of this work comes from NSF PHY-2207697. W.C.H. gratefully acknowledges funding support from NSTC 112-2811-M-001-007 and 113-2811-M-007-008. This work was completed utilizing the Holland Computing Center of the University of Nebraska, which receives support from the UNL Office of Research and Economic Development, and the Nebraska Research Initiative. Manufacturing and characterization analysis were performed at the NanoEngineering Research Core Facility (NERCF), which is partially funded by the Nebraska Research Initiative. The research was performed in part in the Nebraska Nanoscale Facility: National Nanotechnology Coordinated Infrastructure and the Nebraska Center for Materials and Nanoscience (and/or NERCF), which are supported by the National Science Foundation under Award ECCS: 2025298, and the Nebraska Research Initiative.

* Email: waynehuang@mx.nthu.edu.tw
† Email: hbatelaan2@unl.edu

REFERENCES

[1] L. Kronik and Y. Shapira “Surface photovoltage phenomena: theory, experiment, and applications”, Surf. Sci. Rep. 37, 1 (1999).
[2] R. Chen, C. Ni, J. Zhu, F. Fan, and C. Li, “Surface photovoltage microscopy for mapping charge separation on photocatalyst particles”, Nat. Protoc. 19, 2250 (2024).
[3] B.J. Kampherbeek, M.J. Wieland, A. van Zuuk, and P. Kruit, “An experimental setup to test the MAPPER electron lithography concept”, Microelectron. Eng. 53, 279 (2000).
[4] J. R. Anglin, J. P. Paz, and W. H. Zurek, “Deconstructing decoherence”, Phys. Rev. A 55, 4041 (1997).
[5] A. O. Caldeira and A. J. Leggett, “Quantum tunnelling in a dissipative system”, Ann. Phys. 149, 374 (1983).
[6] E. Najafi and A. Jafari, “Ultrafast imaging of surface-exclusive carrier dynamics in silicon”, J. Appl. Phys. 125, 185303 (2019).
[7] W. C. Huang, R. Bach, P. Beierle, and H. Batelaan, “A low-power optical electron switch”, J. Phys. D: Appl. Phys. 47, 085102 (2014).
[8] R. J. Englund et al., “Dielectric laser accelerators”, Rev. Mod. Phys. 86, 1337 (2014).
[9] M. Krüger, M. Schenk, and P. Hommelhoff, “Attosecond control of electrons emitted from a nanoscale metal tip”, Nature 475, 117 (2011).
[10] D. Nabb, J. Kuttruff, L. Stolz, A. Ryabov, and P. Baum, “Attosecond electron microscopy of sub-cycle optical dynamics”, Nature 619, 63 (2023).
[11] A. Feist, K. E. Echternkamp, J. Schauss, S. V. Yalmin, S. Schäfer, and C. Ropers, “Quantum coherent optical phase modulation in an ultrafast transmission electron microscope,” Nature 521, 200 (2015).
[12] P. Sonnentag and F. Hasselbach, “Measurement of decoherence of electron waves and visualization of the quantum-classical transition”, Phys. Rev. Lett. 98, 200402 (2007).
[13] N. Kerker, R. Ropke, L. M. Steinert, A. Poosh, and A. Stibor, “Quantum decoherence by Coulomb interaction”, New J. Phys. 22, 063039 (2020).
[14] P. J. Beierle, L. Zhang, and H. Batelaan, “Experimental test of decoherence theory using electron matter waves”, New J. Phys. 20, 113030 (2018).
[15] Z. Chen and H. Batelaan, “Dephasing due to semiconductor charging masks decoherence in electron-wall systems”, Europhys. Lett. 129, 40004 (2020).
[16] Q. Liu, C. Chen, and H. Ruda, “Surface photovoltage in undoped semi-insulating GaAs”, J. Appl. Phys. 74, 7492 (1993).
[17] M. D. Sturgeon, “Optical absorption of Gallium Arsenide between 0.6 and 2.75 eV”, Phys. Rev. 129, 2835 (1963).
[18] J. R. Hook and H. E. Hall, “Solid state physics”, 2nd ed., p.160, Wiley, Chichester (1991).
[19] H. R. Philipp and H. Ehrenreich, “Optical properties of semiconductors”, Phys. Rev. 129, 1550 (1963).
[20] M. Milanova, A. Mintairov, V. Rumyansev, and K. Smekalin, “Spectral characteristics of GaAs solar cells grown by LPE”, J. Electron. Mater. 28, 35 (1999).
[21] J. Lagowski, P. Edelman, and A. Morawski, “Non-contact deep level transient spectroscopy (DLTS) based on surface photovoltage”, Semicond. Sci. Technol. 7, A211 (1992).
[22] G. M. Martin, “Optical assessment of the main electron trap in bulk semi-insulating GaAs”, Appl. Phys. Lett. 39, 747 (1981).
[23] K. Germanova and Ch. Hardalov, “EL2 deep level in sub-bandgap surface photovoltage spectra in GaAs bulk crystals”, Appl. Phys. A 43, 117 (1987).