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Title:

Spin Hall-driven photocurrent in nonmagnetic single metals

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Abstract

Nonlinear phenomena in condensed matter physics have been explored extensively in exotic materials, including materials with broken inversion symmetry, topological materials, metamaterials, and quantum wells. However, conventional nonmagnetic transition metals have largely been overlooked in this context because of their perceived relative simplicity. Here, we reveal the unexplored potential of nonmagnetic single-element metals by focusing on one type of nonlinear phenomenon: photocurrent generation induced by spin accumulation due to the spin Hall effect at the material edges. Distinctive behavior at the material edges, where the spatial symmetry is intrinsically broken, leads to observation of the nonlinear phenomenon. The resulting photocurrent is identified as a linear injection current and is associated with both time-reversal symmetry breaking and space-inversion symmetry breaking. This observation challenges the prevailing notion that nonlinear phenomena are exclusively observed in more exotic materials. We perform scanning photovoltage microscopy on Pt, W, Cu, and perpendicular magnetic anisotropy films to verify and characterize the observed linear injection current experimentally, thus confirming the crucial roles of spin accumulation and edge asymmetry in photocurrent generation. This finding provides a comprehensive understanding of linear injection currents realized during symmetry violation and is promising for development of spin- and symmetry-based techniques.

Nonlinear phenomena from nonlinear optics¹⁻³ to nonreciprocal transport processes⁴⁻⁷ and the nonlinear Hall effect⁸⁻¹⁰ are prevalent in systems with broken symmetry in condensed matter physics. These nonlinear phenomena have been observed in various exotic materials, including quantum wells^{11,12}, topological materials^{13,14}, synthetic superlattices¹⁵⁻¹⁷, metamaterials^{18,19}, and broken inversion symmetry materials^{4,9,20}, which indicates the potential for use of these materials in deciphering the spatial symmetry and magnetic states of systems. Among these nonlinear phenomena, photocurrent generation based on nonlinear optical processes caused by the electronic band structures of solids, as opposed to the typical use of a p-n junction, is attracting research interest². The ferroelectric domains of noncentrosymmetric two-dimensional (2D) SnS were recently observed via photocurrent mapping²¹ and this distinctive response to the system symmetry illustrates the recognizable convenience of this phenomenon. In this process, the system symmetry determines the relationship between the photocurrent type (shift current or injection current) that results from irradiation of the material and the irradiating light polarization (linear or circular)^{22,23}. One intriguing variant of this photocurrent, the linear injection current, occurs when the time-reversal symmetry and the space-inversion symmetry are broken simultaneously²²⁻²⁴. This phenomenon, which is closely associated with symmetry violation, can thus potentially serve as a tool to investigate the spatial symmetry and magnetic states of systems. This type of photocurrent has been observed in a 2D CrI₃ magnet²⁵, a layered polar BiTeI semiconductor²⁶, and a metamaterial with perpendicular magnetic anisotropy (PMA) and an artificial noncentrosymmetric lattice¹⁹.

Typically, the research into nonlinear phenomena focuses on exotic materials because their unusual characteristics, which include topologically nontrivial band structures, inversion symmetry breaking, and antiferromagnetic spin orders, are integral to manifestation of these nonlinear effects. Notably, the majority of these studies tend to overlook conventional single-element transition metals. This may be the result of the prevailing perception that these 'ordinary' materials lack the complexity required to cause these phenomena to occur. Therefore, our study explores the overlooked potential of simple, nonmagnetic transition metals to host such phenomena. In particular, our attention is drawn to the ability of the linear injection current to probe broken localized symmetry conditions and thus shed light on previously uncharted behavior within these conventional materials.

Nonmagnetic metals under an applied electric current can display local spin accumulation caused by the spin Hall effect (SHE). This spin accumulation, which acts as a localized magnetization, has the capacity to violate the time-reversal symmetry in spatially localized regions. These single-element metals do not inherently break the time-reversal or space-inversion symmetries, but do display distinct behavior at their edges, where the spatial symmetry is intrinsically disrupted. Therefore, even nonmagnetic single-element metals are expected to demonstrate nonlinear phenomena in their edge regions because the situation can cause the time-reversal and space-inversion symmetries to break.

In this study, we investigate a specific type of nonlinear phenomenon: photocurrent generation

induced by spin accumulation due to the SHE at the edge regions of conventional nonmagnetic single-element metals, including platinum (Pt), tungsten (W), and copper (Cu). Furthermore, by comparing our results with those obtained from a PMA film, we confirm that the observed current is a linear injection current caused by spin accumulation. The occurrence of this phenomenon within these conventional metals is groundbreaking and inherently challenges the notion that nonlinear phenomena are exclusive to exotic materials. We aim to reinforce the potential use of the linear injection current as a comprehensive probe for investigation of material symmetry and topology, thereby opening up previously unexplored avenues in the realm of nonlinear phenomena.

The linear injection current in the μ -direction^{22,24} is expressed as

$$j^\mu = \sigma^{\mu;\nu\lambda} E^\nu E^\lambda \quad (1)$$

$$\sigma^{\mu;\nu\lambda} = \frac{-\pi q^3}{\gamma} \int \frac{d\mathbf{k}}{(2\pi)^d} \sum_{a \neq b} \Delta_{ab}^\mu g_{ab}^{\mu;\nu\lambda} f_{ab} \delta(\hbar\omega - \epsilon_{ba}), \quad (2)$$

where E^ν (E^λ) is the ν (λ)-polarized electric field; q , $\hbar$, γ , and ω are the elementary charge, the Dirac constant, the scattering rate, and the angular frequency of light, respectively; Δ , g , and f are the group velocity difference, the quantum metric, and the Fermi-Dirac distribution function, respectively; and ϵ_{ba} is the energy difference, which has been predicted and observed in materials in which the entire bulk system breaks both the time-reversal symmetry and the inversion symmetry^{19,22,23} (Fig. 1a). In contrast to the photocurrents found in exotic materials, which require bulk symmetry breaking, this study investigates photocurrents that arise from localized symmetry breaking in their edge regions, which represent a feature occurs in all realistic systems. The photoconductivity in equation (2) shows a specific relationship with the linear polarization with consideration of the z -direction magnetization:

$$\sigma_z^{x;xx} = -\sigma_{-z}^{x;xx} = \sigma_{-z}^{x;yy} = -\sigma_z^{x;yy}. \quad (3)$$

As shown in Fig. 1b, even without the absence of the inversion symmetry, the edges of materials will break this symmetry spontaneously. Consequently, linear injection current generation in the edge regions is expected when the time-reversal symmetry is broken by either magnetization or a magnetic field. If the system retains its time-reversal symmetry, then the linear injection current is not expected to occur. However, with the spin accumulation caused by the SHE, a scenario can arise where the edge region alone violates both the inversion and time-reversal symmetries, even if the entire bulk does not; this can potentially lead to emergence of linear injection currents at the material's edge (Fig. 1c).

In the case of inversion symmetry-preserving ferromagnets, the direction of the linear injection current at the opposing edges of the material is antiparallel because the two edges have opposite symmetry-breaking directions but the same magnetization direction (Fig. 1b). Conversely, for nonmagnetic metals with spin accumulation, the photocurrent direction remains consistent across both edges because their spin accumulation directions are opposite to each other (Fig. 1c). It is important to note here that the linear injection current direction is dependent not only on the system's symmetry,

but also on the material's intrinsic electronic properties. In addition, a 90° rotation of the linear polarization angle could cause reversal of the current direction, as indicated by equation (3).

In the experiment, we fabricated Hall bar structures using Pt, W, and Cu. All materials were deposited on SiO_2 substrates using an RF magnetron sputtering system (ANELVA, SPC-350). In the cases of W and Cu, a 3-nm-thick SiO_2 capping layer was deposited sequentially in the same sputtering chamber. The Hall bar structures were designed with a width of $10\ \mu\text{m}$. A Hall bar structure with a PMA film was also prepared using the sputtering system. A $[\text{Pt}(3.8\ \text{nm})/\text{Co}(0.8\ \text{nm})]_{10}$ superlattice was deposited on a SiO_2 substrate. The W and PMA channels were fabricated for the control experiments. The W channel was selected because it has the opposite sign for its spin Hall angle when compared with Pt. This allowed us to investigate the relationship between the photocurrent and spin accumulation further. The PMA channel was used to perform an additional control experiment because it breaks the time-reversal symmetry naturally as a result of its magnetization, which then allowed us to examine the role of spin imbalance in the observed photovoltage without having to apply a current. The deposition rates were 3.8 nm/min at 50 W for Pt, 1.5 nm/min at 100 W for W, 4.1 nm/min for Cu, and 0.8 nm/min at 20 W for Co. Temperature of substrates were set to room temperature during the deposition.

To investigate the linear polarization-induced photocurrent in localized areas, scanning photovoltage microscopy^{27,28} is used. We used a continuous-wave 633 nm laser. The linear polarization angle of the laser light is controlled via the $\pi/2$ mode of a photoelastic modulator (PEM) and is varied periodically from parallel to orthogonal with respect to the direction of the metal channels at a frequency of 100 kHz. Prior to light incidence on the PEM, the linear polarization was tilted by 45° with respect to the vertical axis of the PEM. During half-wave plate modulation of the PEM, the linear polarization changed periodically from 45° to -45° and vice versa, where these angles corresponded to the parallel and orthogonal angles with respect to the direction of the metal channels, respectively. During modulation of the polarization angle, the PEM does not change the light intensity, and the thermal fluctuations at the same frequency as the modulation are suppressed. The laser light is focused and is normally incident on the metal channel when a current is applied to induce spin accumulation due to the SHE. The spot diameter was approximately $1\ \mu\text{m}$. We used a lock-in technique to measure the voltage drop caused by the photocurrent across the channel. The detected voltage is proportional to the difference between the photovoltages produced by linearly polarized illumination oriented parallel and orthogonal to the channel direction, and we call this voltage the angle-dependent photovoltage V_{ADP} . Use of second harmonic detection with the PEM enables suppression of a background signal that occurs because of the thermoelectric effect and the contribution of the circular polarization. All measurements were carried out at room temperature.

Figure 2 shows the photocurrent mapping results when a current is applied to the Pt channel. In Fig. 2a, a current of 10 mA is applied and shows that the V_{ADP} signals occur exclusively at the channel edges and have the same sign at both edges. At these edges, the spatial inversion symmetry is disrupted,

and spin accumulation then arises because of the SHE. When a photocurrent scan is performed without application of a current, the signal vanishes (Fig. 2b). Although the edges inherently violate the inversion symmetry, the absence of a signal without current application indicates that the signal obtained cannot be attributed exclusively to disruption of the spatial inversion symmetry. When the direction of the applied current is reversed, the signal is only obtained at the edges in a manner similar to the case with the positive bias, but the sign of the signal is reversed in this case (Fig. 2c). In the cases of positive and negative bias, the directions of the spins that accumulate on the edges are opposite to each other, thus indicating that the sign change in V_{ADP} originates from spin accumulation in opposite directions. Subsequently, the experiment was repeated with the sample being tilted by 90° . The photocurrent results remained consistent in this case, with the signal only being obtained at the edges when a current was applied (Fig. 2d and f). However, by focusing on the signal's sign, it was found that the 90° tilt caused the sign to reverse. This 90° rotation thus alters the relative arrangement between the edge region and the linearly polarized light, indicating that the photocurrent obtained is also influenced by the angle between the direction of the sample edge region and the linear polarization. These results demonstrate that both the current-induced spin accumulation and the breaking of the spatial inversion symmetry in the edge region are essential for photocurrent generation. This photocurrent is attributed to the linear injection current and indicates successful observation of the linear injection current caused by spin accumulation in the edge region. It should be noted that the symmetry of the reported linear injection currents observed in systems with broken symmetry and spin imbalance¹⁹ align well with the present results.

To investigate the contribution of the spin accumulation further, the same experiment was conducted when using β -W as a material replacement for Pt. β -W, which is a material with a negative spin Hall angle, is expected to show reversed spin accumulation behavior in the same current application direction when compared with Pt. Upon measurement of V_{ADP} in a W device on a scale similar to that of the Pt device, signals indicating photocurrents were observed at the edges alone, in a manner similar to the Pt case (Fig. 3a and b). Furthermore, the signs at the two edges were consistent. Comparison of these results with the Pt measurements under the same conditions in Fig. 2a and c confirmed that the sign of V_{ADP} in W had reversed, thus strongly suggesting that the inversion of the linear injection current direction is due to the altered spin accumulation sign. These results clearly rule out the effects of current-induced magnetic fields that could break the time-reversal symmetry at the edge regions of the samples because their directions remain consistent under the same applied current direction; they also indicate that breaking of the time-reversal symmetry due to the current-induced spin accumulation plays an important role in the observed photocurrent. With regard to the other possible origin of the pseudo-signal, the occurrence of the thermoelectric effect due to beam shifting by the PEM was recently indicated²⁹. This effect can also be excluded in the case of our measurement system because of the sign of the signal at both edges and the angular dependence of the channel (see also the

Supplemental Material I)³⁰.

These results demonstrate that the measured photocurrent is the linear injection current derived from spin accumulation at the edge region, thus making it an effective way to read spin states. Furthermore, to examine the relationship between spin imbalance and the linear injection current in more detail, similar experiments were conducted using a PMA film. The PMA was implemented using Pt/Co superlattices that exhibited out-of-plane magnetization at room temperature. By measuring the transverse conductivity while switching the magnetization using an external magnetic field, the presence of a film with out-of-plane magnetization at room temperature was confirmed (see also the Supplemental Material, Fig. S1)³⁰. Upon measurement of the photocurrent in each magnetization direction, signals were obtained at the edge regions only. Because PMA films inherently exhibit spin imbalance without an applied current, we measured the angle-dependent photocurrent I_{ADP} directly, rather than V_{ADP} . Additionally, because the edge signal sign reverses when the magnetization direction is reversed, it can be inferred that the linear injection current, which is affected by both spin imbalance and breaking of the edge spatial inversion symmetry, was observed successfully (Fig. 3c and d). The results differ from those obtained for Pt and W, in that the signal signs at the two edges are distinct. This is because, unlike Pt and W, where the spin accumulation directions at the two edges are opposite because of the SHE (Fig. 1c), PMA films show uniform spin imbalance across the entire film (Fig. 1b). The linear injection current reverses its sign during both spatial inversion and time-reversal operations (inversion of the magnetization direction), indicating a good match between the signal signs obtained and the symmetrical property of the linear injection current. The control experiment with PMA highlights the robustness and versatility of the proposed method because it can detect spin accumulation even in the absence of the applied current. This illustrates the potential of linear injection currents for investigation of a wide range of materials and systems, opening up new opportunities for exploration of the symmetrical properties and electronic structures of materials.

Investigation of the laser intensity and bias current dependences of the V_{ADP} signal is not only essential but also is decisive in establishing the relationship between the linear injection current and the spin accumulation. We measured the magnitude of V_{ADP} at a fixed spot on the Pt channel edge while varying both the laser intensity and the bias current. The results showed a linear dependence on the bias current (Fig. 4a), which was also confirmed by the current reversal shown in Fig. 2. This indicates that the observed signal was induced by the current. A previous study that measured the photocurrents in a broken inversion symmetry material, in which the time-reversal symmetry was broken by an external magnetic field, reported that the photocurrent's magnitude is proportional to the magnitude of the external magnetic field²⁶, and the linear relationship observed in this study in particular supports the notion that this phenomenon can be attributed to spin accumulation. Similarly, we observed a linear dependence of the V_{ADP} on the laser intensity (Fig. 4b). In the low-intensity regime, the linear injection current is proportional to the intensity because it represents a quadratic

effect with respect to the excitation light's electric field. This observation is in full agreement with the expression for the linear injection current in equation (1), thus reinforcing the connection between V_{ADP} and the underlying theory conclusively.

Detection of spin accumulation in a lighter metal, e.g., Cu, which has a small spin-orbit coupling, was demonstrated via the linear injection current measurement method. The results of V_{ADP} mapping are shown in Fig. 5. A clear V_{ADP} voltage was detected at the edge of a single Cu channel, as also observed in Pt and W. This unexpected result is caused by the long spin coherence of Cu. The spin Hall angle of Cu is as low as 0.0032, but its spin diffusion length is much higher than those of heavier metals³¹. Under this condition, if the channel width is sufficiently greater than the spin diffusion length, then a nonnegligible spin accumulation occurs at the Cu edge (see also the Supplemental Material II)^{30, 32-34}. In conventional SHE detection methods, e.g., spin torque ferromagnetic resonance, spin valve, and photovoltage measurements with circular polarization, this type of spin accumulation in light metals with small spin-orbit coupling has hardly been observed previously. The linear injection current, which requires neither a junction with a ferromagnet nor spin-orbit interactions, reflects the violated symmetry condition directly even in a single-element light metal, and thus this previously unobserved spin accumulation can be visualized.

Finally, the technique used in our study offers an alternative method to detect spin accumulation textures in metals. As the conventional technique, the magnetic optical Kerr effect (MOKE) has been used to probe current-induced spin accumulation in metals³⁴⁻³⁶. However, some studies have indicated the absence of a detectable MOKE signal and the existence of spurious signals^{37, 38}. Measurement of the linear injection current gives a symmetry-protected response that reflects the local inversion symmetry and orientation of the accumulated spins, making it possible to detect symmetry breakage within a highly localized region in the metals, which gives a smaller MOKE signal. In this study, we demonstrated the detection of spin accumulation in the xz plane, as shown in Fig. 1. It is anticipated that the proposed method can also detect spin accumulation at other boundaries such as the xy plane, offering an alternative approach to current techniques. For example, y -oriented spin accumulation in the xy plane can be observed using oblique light incidence and optimizing the photocurrent detection direction based on the spin orientation. These modifications highlight the method's versatility in detecting spin accumulation in various spatial configurations. Our technique is thus advantageous for detection of spins accumulated within nanoscale regions in metals. In addition, since time-reversal symmetry breaking is a requirement, this technique may also be able to detect an accumulation of the orbital angular momentum that have recently received a great deal of attention for the application of spin torque induced by the orbital Hall effect.

The underlying physical principle of this discovery lies in the manifestation of nonzero quantum metrics when both the time reversal symmetry and the inversion symmetry are broken. Nonlinear

phenomena stemming from such quantum metrics, including the nonlinear Hall effect^{39,40}, have been attracting attention recently. Nonlinear phenomena driven by quantum metrics, including the results presented in this paper, are independent of the scattering time because of the combined symmetry of the time-reversal and inversion symmetries, making these phenomena attractive probes with suppressed impurity scattering and high sensitivity to band engineering^{22, 41}. Therefore, measurement and mapping of the linear injection current not only enables detection of spin accumulation but also provides a promising method to observe and quantify the spatial distributions of novel physical quantities and modulated band structures.

In summary, our findings confirm that linear injection currents, which are a type of nonlinear phenomenon, can occur even in non-exotic materials, e.g., nonmagnetic single-element metals such as Pt, W, and Cu. We found that current-induced spin accumulation and spatial inversion symmetry breaking at the edge region play essential roles in generation of this photocurrent. In other words, we have observed spin accumulation in the material edge regions successfully using the linear injection current. Experiments using W as an alternative material with a negative spin Hall angle and PMA with spontaneous spin imbalance further supported and highlighted the link between time-reversal symmetry breaking because of spin imbalance and the observed linear injection current. These findings, which provide a comprehensive understanding of the relationship between spin imbalance, the linear injected current, and the edge asymmetry, have important implications for the nonlinear physics field and are promising for the development of spin- and symmetry-based measurement techniques.

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Author contributions

T. N., Y. Y. and Y. A. conceived the experiments. T. N. fabricated samples and collected data. T. N, R. O., R. I., Y. Y., M. S., and Y. A. analyzed results. T.N. and Y.A. wrote the manuscript. All authors discussed the results.

Data availability

Source data for the main paper figures are provided with this paper. Additional data are available from the corresponding authors on request.

Competing interest

The authors declare no competing interests.

Additional information

Supplemental Material is available for this paper at XXX.

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Figure captions

Fig. 1 Schematic of the linear shift current generation process in materials with different symmetries. Green arrows represent the directions of the linear shift currents.

a Ferromagnetic and noncentrosymmetric materials. The color gradient from blue to green represents breaking of the material symmetry. The time reversal symmetry and the inversion symmetry are broken simultaneously in the entire system, and thus the linear shift current occurs in the bulk.

b Ferromagnetic centrosymmetric material. The time reversal symmetry is broken in the entire system, but the inversion symmetry is only broken at the edge region. The linear shift current is generated at the edge region only and the current directions are opposite on the other side of the edge.

c Nonmagnetic centrosymmetric metal with spin accumulation due to the SHE. The time reversal symmetry and the inversion symmetry are broken at the edge region only. The linear shift current is generated at the edge region only, but with the same direction as the current on the other side of the edge.

Fig. 2 Spatial two-dimensional maps of the angle-dependent photovoltage in 10-nm-thick Pt under bias currents of **a** 10, **b** 0, and **c** -10 mA, and the 90°-rotated case under bias currents of **d** 10, **e** 0, and **f** -10 mA. Black dashed lines indicate the channel edges, which were confirmed by reflection measurements that were conducted simultaneously. Black bold arrows indicate the applied bias current direction. The intensity of the incident laser light was 6.4 mW/μm².

Fig. 3 a, b Spatial two-dimensional maps of the angle-dependent photovoltage in 10-nm-thick W under bias currents of **a** 10 and **b** -10 mA. **c, d** Spatial two-dimensional maps of the angle-dependent photovoltage in PMA with **c** positive and **d** negative out-of-plane magnetizations, without a bias current. Black dashed lines indicate the channel edges, as confirmed by reflection measurements conducted simultaneously. Black bold arrows indicate the applied bias current direction. The intensities of the incident laser light were 13 mW/μm² for W and 2.6 mW/μm² for PMA.

Fig. 4 Characterization of the angle-dependent photovoltage. **a.** Bias current dependence. **b.** Laser intensity dependence.

Fig. 5 a, b Spatial two-dimensional maps of the angle-dependent photovoltage in 10-nm-thick Cu, under bias currents of **a** 10 and **b** -10 mA. The intensity of the incident laser light was 6.4 mW/μm².

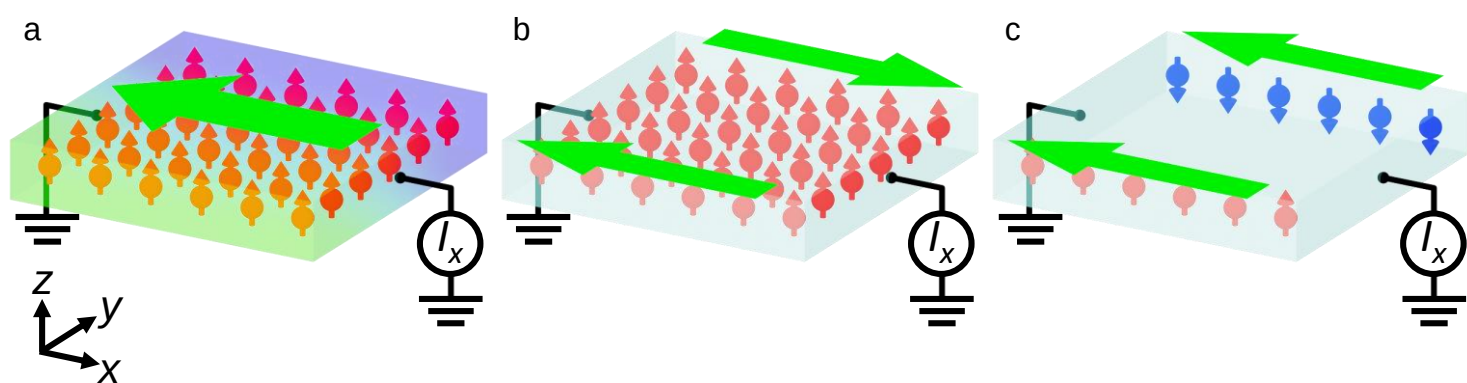


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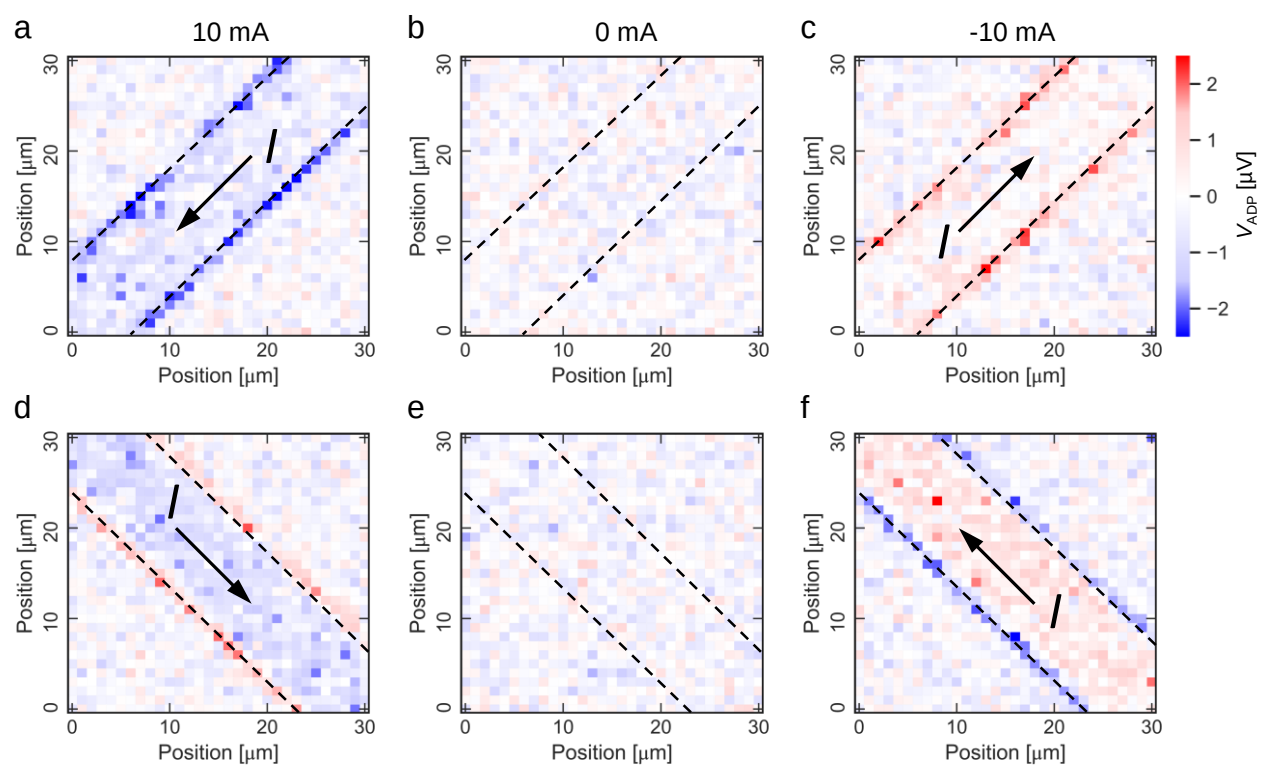


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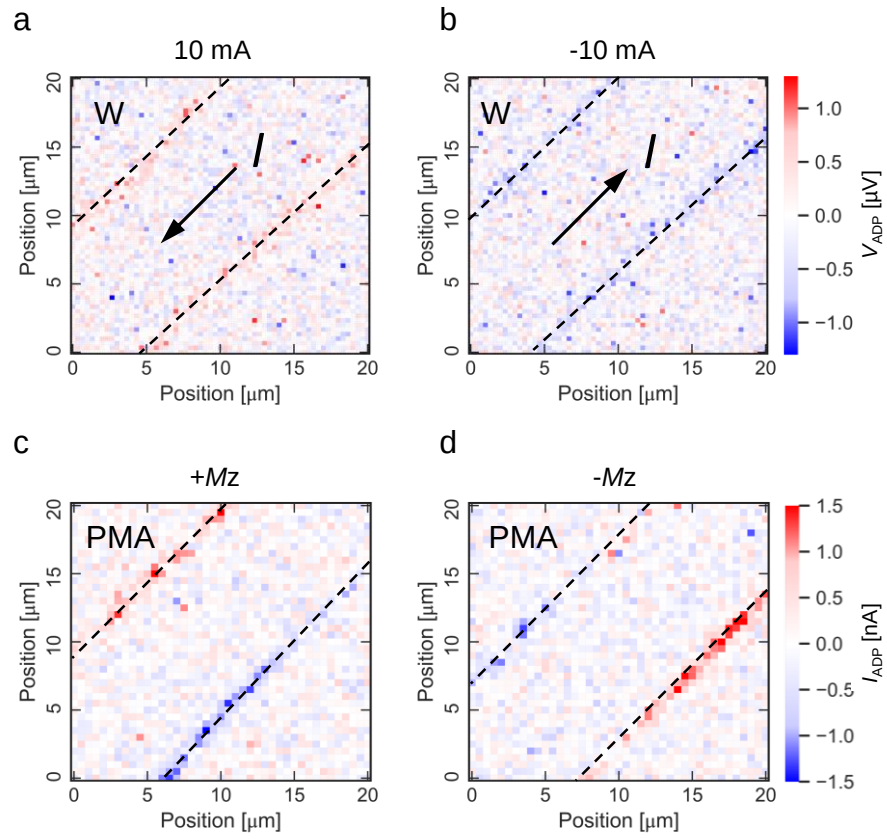


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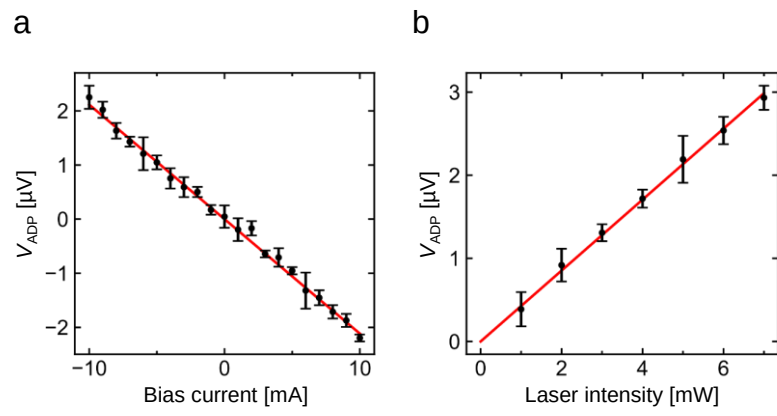


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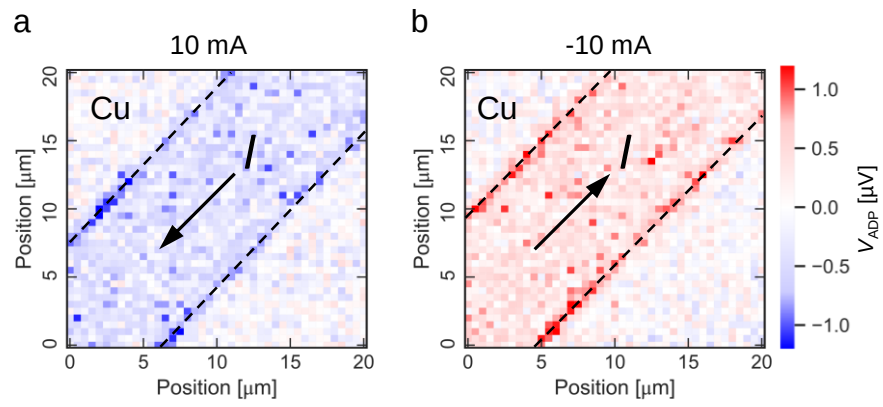


Fig. 5 T. Nishijima et al.,