

Terahertz generation via the inverse orbital Rashba-Edelstein effect at the Ni/CuO_x interface

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Orbitronics provides novel mechanisms to generate terahertz (THz) emission using the orbital angular momentum of electrons. Here, we report a systematic investigation of the THz emission from Ni/CuO_x heterostructures pumped by a femtosecond laser pulse. Despite the very weak spin-orbit coupling of CuO_x, considerable THz radiation from Ni/CuO_x has been observed, with a polarity state that is magnetically controllable and the intensity as much as about 20% of that found in Ni/Pt spintronic THz emitter. We find conclusive evidence that THz radiation stems from the inverse orbital Rashba-Edelstein effect at the Ni/CuO_x interface, which can be manipulated by tuning oxygen-induced orbital hybridization. Our experimental results of Ni thickness-dependent THz emission, combined with theoretical modeling, reveal long-range diffusion-length characteristics of orbital current within the Ni layer. This work enhances the understanding of the THz emission mechanism based on orbital-to-charge conversion, providing guidance for the development of orbitronic THz emitters.

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Spintronic terahertz (THz) emitters based on spin-to-charge conversion (S-C) have attracted tremendous attention as broad-bandwidth, highly efficient, and highly flexible sources of THz radiation [1–4]. A typical spintronic THz emitter is a magnetic heterostructure composed of a ferromagnetic metal (FM) and a nonmagnetic material (NM) with a strong spin-orbit coupling (SOC) [1,5–8]. Ultrafast spin current J_S is generated in the FM layer by femtosecond (fs) laser excitation, then injected into the NM layer through a mixture of ballistic and diffusive (superdiffusive) transport, and is converted into a transient transverse charge current J_C via the inverse spin Hall effect (ISHE) [9–14] or the inverse Rashba-Edelstein effect (IREE) [15,16], leading to THz pulse emission. However, in addition to spin angular momentum (SAM), electrons also possess orbital angular momentum (OAM) [17–19]. Recent studies have shown that the orbital current J_L , excited by the fs laser in the FM layer, can be converted to J_C via the inverse orbital Hall effect in the NM layer with weak SOC, achieving orbitronic THz emission [20,21]. These findings have opened an emerging field of

ultrafast orbitronics, demonstrating the significant potential for generating and controlling THz radiation.

Recently, oxidized light metals such as copper oxide (CuO_x) with negligible SOC have become attractive in orbitronics due to their lower cost compared to heavy metals and the tunability of oxygen-induced orbital hybridization through oxidation levels [22–25]. In FM/CuO_x heterostructures, electrical transport experiments have indicated that nonequilibrium OAM accumulation can be electrically generated by the orbital Rashba-Edelstein effect (OREE) [26–30]. In particular, the orbital torque produced by CuO_x material can be effectively utilized for electrical control of magnetization in the FM layer [31,32]. Conversely, the mechanism of the inverse orbital Rashba-Edelstein effect (IOREE) allows for the conversion of OAM into charge currents at the symmetry-broken interface, independent of SOC. When orbital-polarized electrons are injected into the orbital Rashba-Edelstein interface, the chiral orbital-polarized Fermi surface undergoes a shift by Δk [highlighted by the red arrow in Fig. 1(a)], inducing a nonequilibrium state in the Fermi band and subsequently generating a charge current along the direction of Δk . However, THz emission originating from the IOREE mechanism in FM/CuO_x heterostructures has not yet been explored.

In this work, we report the observation of THz radiation from the Ni/CuO_x bilayer system stemming from the IOREE. The THz intensity is approximately 20% of that of the Ni/Pt spintronic emitter and is influenced by the oxidation level of the Cu layer. A 180° phase shift is observed when the external magnetic field is reversed. To understand the

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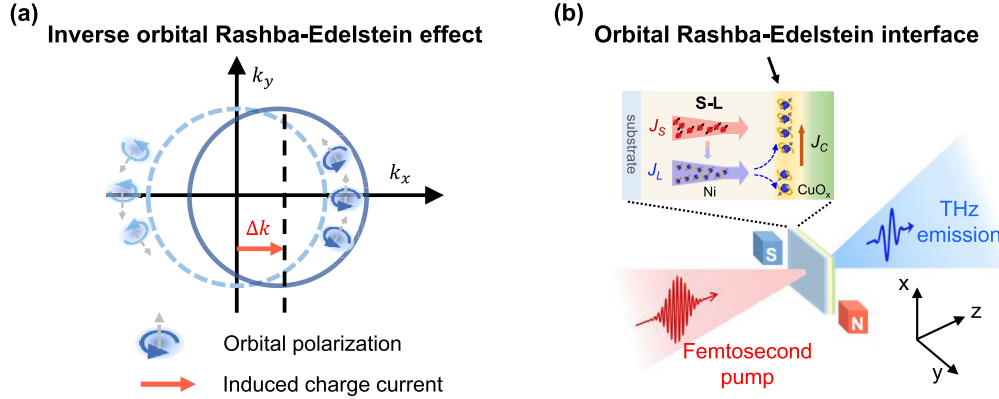


FIG. 1. (a) Illustration of the inverse orbital Rashba-Edelstein effect: Injection of OAM induces an extra population on one of the sides of the Fermi contour, a depletion on the other side, and, therefore, a charge-current density. (b) A sketch for the experimental generation of THz emissions of Ni/CuO_x bilayer samples for substrate side pumping, where the femtosecond laser propagates along the *z* axis and the magnetic field is applied along the *y* axis. The orbit-to-charge conversion occurs at the orbital Rashba-Edelstein interface of Ni/CuO_x.

underlying physics, we developed theoretical models to analyze Ni thickness-dependent THz emission from Ni/CuO_x and Ni/Pt systems. The THz amplitude of the Ni/CuO_x system peaks at a thicker Ni layer compared to Ni/Pt, attributed to the larger orbital diffusion length in Ni. Our findings demonstrate an efficient method for generating orbitronic THz emission and elucidate the IOREE mechanism.

The Ni/Cu stacks were deposited on 0.5-mm-thick Al₂O₃ (0001) substrates by magnetron sputtering at a base pressure of 2×10^{-9} mbar. After deposition, the Al₂O₃/Ni/Cu samples were removed from the vacuum and exposed to the atmosphere at room temperature for 48 h to form naturally oxidized Al₂O₃/Ni/CuO_x samples, and then THz measurements were conducted. Note that the series of samples in our study experienced the exact same natural oxidation process to ensure that the THz measurement results were comparable between different Al₂O₃/Ni/CuO_x samples. A standard THz time-domain spectroscopy setup was utilized to generate and detect the THz pulse waveforms (see more details in Supplemental Material) [33]. As depicted in Fig. 1(b), linearly polarized femtosecond laser pulses (center wavelength of 800 nm, pulse duration of 35 fs, repetition rate of 1 kHz, pump fluence of 0.19 mJ/cm²) were incident from the substrate side to the Ni/CuO_x bilayer along the *z* axis to avoid undesired THz absorption by the substrate. A magnetic field of 1000 Oe was applied along the *y* axis to align the magnetic moment of Ni. The THz signal, with the electric field along the *x* axis, was detected using a 2-mm-thick ZnTe crystal via electro-optical sampling.

The absorption of the femtosecond laser pulses of the Ni layer excites the transport of spin-polarized hot electrons from the Ni into the CuO_x layer [16]. During the transport within the Ni layer, the SAM carried by hot electrons is converted to OAM due to the SOC of Ni. Subsequently, both the spin current (*J_S*) and orbital current (*J_L*) are injected into the CuO_x layer [20]. Considering the weak SOC of CuO_x, the S-C is negligible. However, the orbital accumulation can be converted to the charge current (*J_C*) on a picosecond timescale through IOREE [Fig. 1(b)], generating a THz electric field along the *x* axis. As reported, the oxygen

concentration gradient along the *z* axis in the CuO_x layer is introduced by natural oxidation [26,29,34], and the copper immediately adjacent to the Ni layer is not fully oxidized [22], thus allowing the orbit-to-charge conversion (L-C) by IOREE [23]. To obtain information about the naturally oxidized Cu films in our work, the oxidation levels of the CuO_x layer in the Ni(9)/CuO_x(3) and Ni(9)/CuO_x(7) (numbers indicate thickness in nanometers) samples are characterized by angle-resolved x-ray photoelectron spectroscopy and Auger electron spectroscopy (details in Supplemental Material, S3) [33,35]. It is observed that the CuO_x layer, which is a mixture of CuO, Cu₂O, and Cu components, is not fully oxidized. Nevertheless, an oxidation gradient is formed along the film normal, indicating that the metallic Cu is distributed near the Ni/CuO_x interface.

Figure 2(a) shows the typical THz temporal waveforms from the Ni(9)/CuO_x(3) bilayer sample, obtained by pumping from the substrate side. In fact, THz radiation from a ferromagnetic metal can be generated by ultrafast demagnetization, anomalous Hall effect, and anomalous Nernst effect [36–38]. Consequently, we performed a control experiment on a single ferromagnetic layer, which was covered by a MgO(2)/Ta(2) capping layer where the MgO layer was used to protect the Ni layer from oxidation, and the additional Ta layer was added to prevent the deliquescence of the MgO layer. As depicted in Fig. 2(a), the amplitude of the THz signal of 9-nm-thick Ni is much weaker than that of the Ni(9)/CuO_x(3) sample, while a 180° phase shift is observed. These results strongly suggest that the THz emission mainly originates from the Ni(9)/CuO_x(3) bilayer structure rather than the Ni layer. Figure 2(b) displays the THz signals of Ni(9)/CuO_x(3) under opposite magnetization $\pm M$ of the Ni layer, where the direction of the magnetization $\pm M$ is set by the external magnetic field. Obviously, the polarity of the THz waveform is inverted when *M* is reversed. Based on the IOREE mechanism for THz generation, the relation between the orbital current and the charge current is described by the equation $J_C = \theta_{\text{IOREE}}^{\text{eff}} J_L \times \sigma_L$, where $\theta_{\text{IOREE}}^{\text{eff}}$ and σ_L are the effective L-C efficiency for the Ni/CuO_x heterostructure and the orbital polarization, respectively. *J_L* originates from the spin-to-orbit

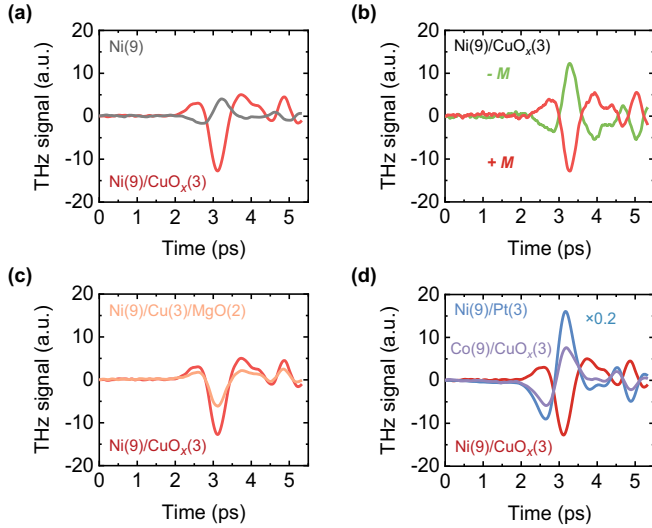


FIG. 2. (a) Typical temporal THz waveforms generated by Ni(9)/CuO_x(3) bilayer and Ni(9) single layer (thickness in nanometers). (b) The THz signal of Ni(9)/CuO_x(3) bilayer inverts with reversal of the sample magnetization $\pm M$. (c) THz emission signals of samples under different oxidation processes, including the natural oxidation for Ni(9)/Cu(3)/MgO(2) and MgO cap-induced oxidation for Ni(9)/Cu(3)/MgO(2). (d) Comparison of THz waveforms of the Ni(9)/CuO_x(3), Co(9)/CuO_x(3), and Ni(9)/Pt(3) bilayer samples. Note the rescaling factor for Pt.

conversion (S-L) due to the SOC of the Ni layer, and the over half-filled d -shell electron configuration of Ni results in σ_L being in the same direction as spin polarization σ_S , which directly depends on the magnetization orientation of Ni. When the applied magnetic field is reversed, the direction of σ_S , σ_L , and J_C also reverses, resulting in a phase change of 180° in the output THz waveform.

In FM/CuO_x systems, the interfacial orbital hybridization, which determines the strength of OREE/IOREE, can be modulated by tuning the oxidation level of the Cu layer [28,29,39,40]. To identify whether the IOREE at the CuO_x interface plays an important role in Ni/CuO_x THz emission, we adjusted the oxidation level of the copper layer by introducing an adjacent oxide capping layer, MgO. The pronounced difference in resistance between copper and its oxides allowed us to evaluate the oxidation level of the copper layer in the Ni(9)/CuO_x(3) sample and Ni(9)/Cu(3)/MgO sample through resistance measurements [29]. Our results indicate that a 48-h air exposure leads to a higher oxidation level of the copper layer in Ni(9)/CuO_x(3), as detailed in Supplemental Material, S2 [33]. Different oxidation processes do not affect the polarity of the THz waveform but strongly affect the intensity of the THz emission, as shown in Fig. 2(c). The peak-to-peak amplitude of the THz waveform for Ni(9)/Cu(3)/MgO(2) sample is 50% smaller than that of the Ni(9)/CuO_x(3) sample. The L-C efficiency can be expressed by $\theta_{\text{IOREE}}^{\text{eff}} = (2e/\hbar)\rho\sigma_{\text{OH}}^{\text{CuO}_x}$, where ρ and $\sigma_{\text{OH}}^{\text{CuO}_x}$ are electrical resistivity and effective orbital Hall conductivity of the CuO_x layer, respectively [41]. Compared to oxidation induced by the MgO cap layer, the natural oxidation process achieves a higher oxidation level, leading to the larger ρ of the CuO_x layer for the Ni(9)/CuO_x(3) sample. Furthermore, a higher level of oxidation enhances

orbital hybridization, resulting in a larger $\sigma_{\text{OH}}^{\text{CuO}_x}$ [28,42]. Therefore, the larger $\theta_{\text{IOREE}}^{\text{eff}}$ of Ni(9)/CuO_x(3) will contribute to the larger intensity of the THz emission.

To compare with conventional spintronic THz emission, we next investigated the THz emission from Ni(9)/CuO_x(3), Co(9)/CuO_x(3), and Ni(9)/Pt(3). As shown in Fig. 2(d), the THz intensity of Ni/CuO_x is as large as about 20% of that of the Ni/Pt reference sample. In addition, the THz polarity of the Ni/CuO_x sample is opposite to that of the Co/CuO_x and Ni/Pt samples, indicating that the direction of the charge current generating THz emission in Ni/CuO_x is opposite to that in Co/CuO_x and Ni/Pt. Due to the strong SOC of the Pt layer, S-C in Ni/Pt is dominated by the bulk ISHE of Pt, described by the equation $J_C = \theta_{\text{ISHE}} \cdot J_S \times \sigma_S$, where the θ_{ISHE} indicates the S-C efficiency of Pt and σ_S is the spin polarization. As reported, the sign of the spin-to-orbit conversion efficiency in both Ni and Co is positive [43], where a positive sign means the same polarization of the spin current and the generated orbit current. Given the identical polarization of the spin currents in the Ni and Co layer under the same magnetic field, the polarization of the orbital currents generated from Co and Ni possesses the same sign. Along with a positive θ_{ISHE} in the Pt layer [43], the THz signal polarity observed in our samples suggests negative $\theta_{\text{IOREE}}^{\text{eff}}$ of Ni/CuO_x, while positive $\theta_{\text{IOREE}}^{\text{eff}}$ of Co/CuO_x. The opposite sign of $\theta_{\text{IOREE}}^{\text{eff}}$ is believed to result from band-structure modifications caused by different FM layers [20,40,44].

Theoretical descriptions of the spintronic THz emission process involving the pump-pulse absorption, spin generation, and S-C have been well established and applied in different analyses [45]. However, a theoretical model based on orbitronic THz emission, which is necessary to understand THz emission via the OAM channel, is absent. Therefore, we build a model considering both spintronic and orbitronic contributions, and the THz electrical field can be expressed as follows:

$$E_{\text{THz}} = E_{\text{THz}}^S + E_{\text{THz}}^L, \quad (1)$$

where E_{THz}^S and E_{THz}^L represent the spintronic and orbitronic components, respectively. Based on previous studies [1,45], we adopt a theoretical model to describe film-thickness dependent THz emission, where $E_{\text{THz}}^{S(L)}$ can be expressed as

$$E_{\text{THz}}^{S(L)} \propto \frac{P_{\text{abs}}}{t_{\text{FM}} + t_{\text{NM}}} \frac{j_{S(L)} Z_0}{1 + n + Z_0(\sigma_{\text{FM}} t_{\text{FM}} + \sigma_{\text{NM}} t_{\text{NM}})} \times \tanh\left(\frac{t_{\text{NM}}}{2\lambda_{S(L)}^{\text{NM}}}\right) \quad (2)$$

where P_{abs} is the laser power absorbed by the FM/NM bilayer, $t_{\text{FM(NM)}}$ is the thickness of the FM (NM) layer, $j_{S(L)}$ is the spin (orbital) current density, n is the refractive index of the substrate, Z_0 is the impedance of the vacuum, $\sigma_{\text{FM(NM)}}$ is the electrical conductivity of the FM (NM) layer, and $\lambda_{S(L)}^{\text{NM}}$ is the spin- (orbital) diffusion length of the NM layer. We note that the sign of $j_{S(L)}$ represents the spin- (orbital) polarization direction, and the positive (negative) sign indicates that the polarization direction is parallel (antiparallel) to the external magnetic field, respectively. This model comprehensively describes the generation, transport, and accumulation of spin

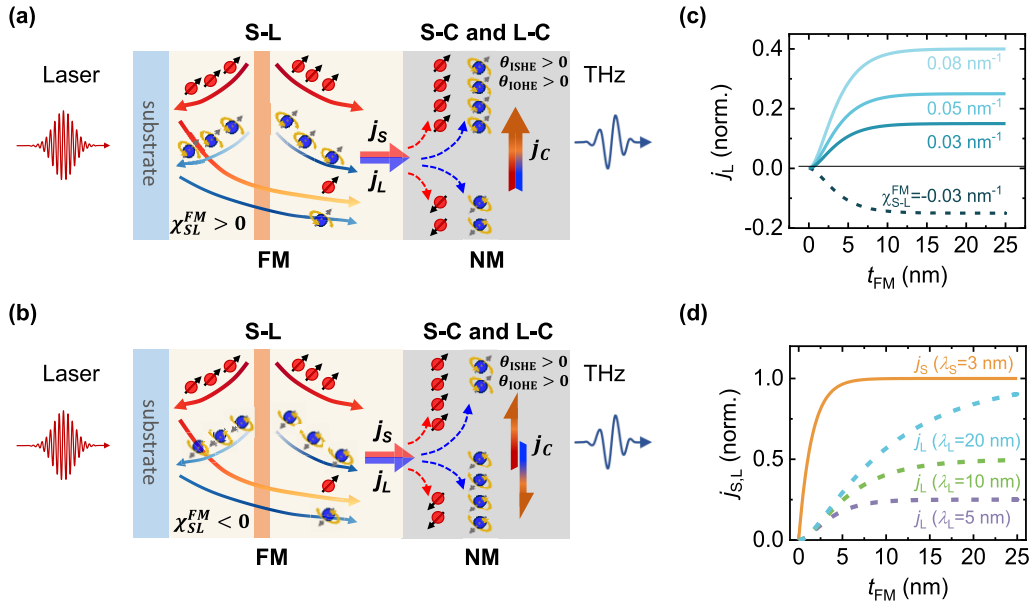


FIG. 3. (a), (b) Schematics of the S-L, S-C, and L-C conversions in the FM/NM systems with the spin- and orbital profile in the FM layer possessing positive and negative S-L conversion efficiency, respectively. (c) Calculated orbital currents at the FM/NM interface as a function of t_{FM} with different values. (d) FM layer-thickness dependence of calculated spin- and orbital currents at the FM/NM interface, where the orbital diffusion length varies from 5 to 20 nm.

(orbital) current, along with the optical properties of FM/NM bilayer samples. Note that although the model is based on the conversion of a flow of angular momentum into a charge current, it remains applicable to the Ni/CuO_x system, where the accumulation of OAM is converted into a charge current (see details in Supplemental Material, S4) [33]. However, the orbital pumping yield through the S-L process in the FM layer remains unclear and warrants further investigation. Therefore, we explored the dynamics of SAM and OAM in the FM layer during the THz emission process.

As shown in Fig. 3(a), the laser pulse illuminates throughout the FM layer and excites spin current in opposite directions at each thin region [46]. The propagation of generated spin current flows is accompanied by an exponential attenuation with a spin-diffusion length λ_S . Considering the interfacial reflections and absorptions (see details in Supplemental Material, S4) [33], the final output spin current of the FM layer can be expressed as

$$j_S = D\lambda_S(1 - e^{-2(t_{FM}-t_0)/\lambda_S}), \quad (3)$$

where D is the spin current density of an infinitesimal region of the FM layer excited by the laser pulse in the FM layer, and t_0 is the dead-layer thickness of FM material. We treat the generation of orbital current as a secondary effect that follows the generation of the spin current in the FM layer with strong SOC [47]. The transport of the generated orbital current can be characterized by an orbital diffusion length λ_L (see Supplemental Material, S4 for a detailed derivation), and the final output orbital current density of the FM layer can be expressed as

$$j_L = \frac{\chi_{SL}^{FM} D \lambda_L \lambda_S}{\lambda_L - \lambda_S} [\lambda_L(1 - e^{-2(t_{FM}-t_0)/\lambda_L}) - \lambda_S(1 - e^{-2(t_{FM}-t_0)/\lambda_S})], \quad (4)$$

where χ_{SL}^{FM} represents the efficiency of S-L within an infinitesimal thickness of the FM layer. The magnitude of χ_{SL}^{FM} is related to the SOC strength, and its sign determines the OAM polarization direction. For $\chi_{SL}^{FM} > 0$, the polarization direction of the orbital current is the same as that of the spin current during the process of S-L in the FM layer. With both the spin Hall angle and orbital Hall angle positive ($\theta_{SH}^{NM} > 0$ and $\theta_{OH}^{NM} > 0$), the charge currents obtained by L-C and S-C possess the same direction in the NM layer, as shown in Fig. 3(a). For $\chi_{SL}^{FM} < 0$ [Fig. 3(b)], the polarization direction of the orbital current is opposed to that of the spin current, resulting in compensating the output charge currents in the NM layer.

To get a quantitative description of the S-L process, we calculated j_L at the FM/NM interface as a function of the t_{FM} under χ_{SL}^{FM} with different values shown in Fig. 3(c). The parameters $\lambda_S = 3$ nm, $\lambda_L = 5$ nm, and $t_0 = 0$ nm were used appropriately in Eq. (4). As expected, the sign of χ_{SL}^{FM} determines the sign of j_L , which represents the direction of orbital polarization. Moreover, the absolute value of j_L enhances with the increase of t_{FM} and then reaches saturation. The larger the value of χ_{SL}^{FM} , the greater the magnitude of j_L . In order to compare the characteristics of spin and orbital currents, the FM layer-thickness dependence of calculated spin and orbital currents are depicted in Fig. 3(d), where χ_{SL}^{FM} and λ_S are fixed as constants of 0.05 nm^{-1} and 3 nm, respectively. λ_L ranges from 5 to 20 nm, and $t_0 = 0$ nm. As the thickness of the FM layer increases, j_S rapidly increases and saturates at $t_{FM} = 5$ nm, a typical behavior due to the short spin-diffusion length. The generated orbital currents with different λ_L remain relatively consistent within the initial 3-nm span, a result of the uniform χ_{SL}^{FM} . Notably, when $t_{FM} > 3$ nm, j_L strongly depends on λ_L . The increase of λ_L not only enlarges the magnitude of j_L , but also causes j_L to trend towards saturation at greater

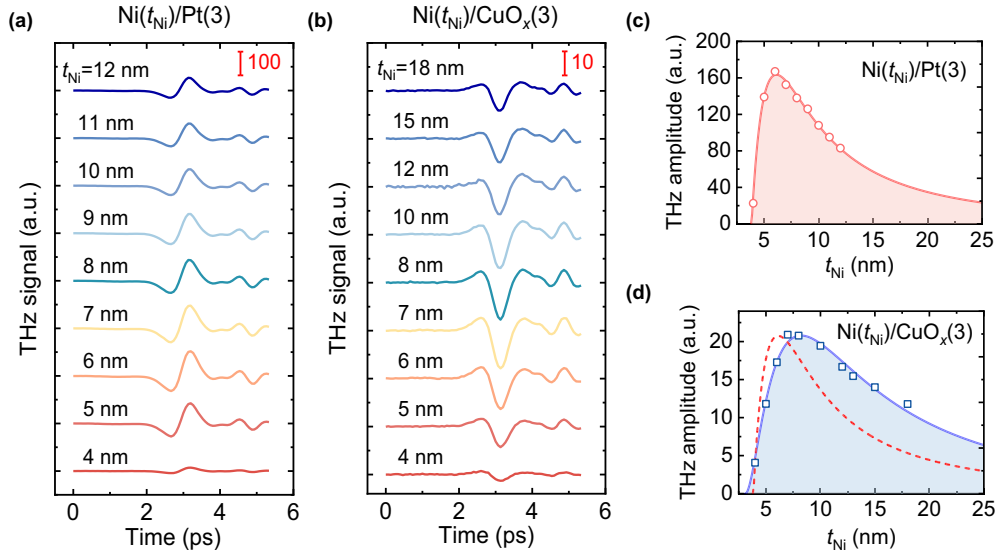


FIG. 4. THz emission signals of (a) $\text{Ni}(t_{\text{Ni}})/\text{Pt}(3)$ and (b) $\text{Ni}(t_{\text{Ni}})/\text{CuO}_x(3)$ samples. THz amplitude dependence on Ni layer thickness for (c) $\text{Ni}(t_{\text{Ni}})/\text{Pt}(3)$ spintronic THz emitters and (d) $\text{Ni}(t_{\text{Ni}})/\text{CuO}_x(3)$ orbitronic THz emitters. Symbols represent experimental data, while lines correspond to theoretical results. The red dashed line in (d) shows the fitting results of $\text{Ni}(t_{\text{Ni}})/\text{Pt}(3)$ samples scaled by a factor of 0.12, demonstrating a significant deviation from the experimental data of $\text{Ni}(t_{\text{Ni}})/\text{CuO}_x(3)$ samples.

t_{FM} . Therefore, the investigation of FM layer-thickness dependence provides an effective means to quantitatively analyze the diffusion length, thereby distinguishing the contributions from spintronics and orbitronics.

We conducted further experiments for the $\text{Ni}(t_{\text{Ni}})/\text{Pt}(3)$ spintronics THz emitter and $\text{Ni}(t_{\text{Ni}})/\text{CuO}_x(3)$ orbitronics THz emitter, which had a fixed Pt or CuO_x thickness but different Ni layer thickness ($t_{\text{Ni}} = 4 \sim 18$ nm, which is thicker than the dead layer [48,49]). As shown in Figs. 4(a) and 4(b), the THz pulse is obtained in all samples, and its amplitude varies with t_{Ni} . Figures 4(c) and 4(d) summarize the extracted THz peak-to-peak amplitude for Ni/Pt and Ni/ CuO_x , respectively, as a function of the thickness of the Ni layer. With the increase of t_{Ni} , the THz amplitude of both Ni/Pt and Ni/ CuO_x systems increases significantly at first, then undergoes a smooth decrease after reaching a peak. The obvious difference is that the Ni thickness corresponding to the maximum THz amplitude of the Ni/ CuO_x system is greater than that of the Ni/Pt system. A theoretical analysis of the experimental results was conducted to further quantify the thickness dependence of the THz amplitude. We simulated the $E_{\text{THz}}^S - t_{\text{Ni}}$ relation for Ni/Pt based on Eqs. (2) and (3), and the $E_{\text{THz}}^S - t_{\text{Ni}}$ relation for Ni/ CuO_x based on Eqs. (2) and (4). In Figs. 4(c) and 4(d), the red and blue curves represent the fitting results for the Ni/Pt and Ni/ CuO_x samples, respectively, showing good agreement with the measurements. For clearer comparison, the fitting results of Ni/Pt were scaled by 0.12 and plotted as a dashed red line in Fig. 4(d). During the fitting process, we adopted the constant parameter $n = 3.2$, $Z_0 = 377 \Omega$ [50], and assumed the conductivity $\sigma_{\text{Ni}}(t_{\text{Ni}}) = \sigma_{\text{Ni}}^{\text{bulk}} / (1 + C_{\text{Ni}}/t_{\text{Ni}})$, where $\sigma_{\text{Ni}}^{\text{bulk}} = 3.3 \times 10^6 \Omega^{-1} \text{ m}^{-1}$ and $C_{\text{Ni}} = 4.6 \times 10^{-9} \text{ m}$ are extracted by fitting the σ_{Ni} measured for different values of t_{Ni} (Fig. S2) [33]. For Ni/Pt spintronics THz emitter, the spin diffusion in Pt has been widely investigated [51], and the fitting parameter $\lambda_S^{\text{Pt}} = 1.5$ was selected [41]. The measured conductivity of 3-nm-thick

Pt film was $\sigma_{\text{Pt}} = 1.8 \times 10^6 \Omega^{-1} \text{ cm}^{-1}$. From the fitting, we obtain the spin-diffusion length in Ni $\lambda_S^{\text{Ni}} = 3.3 \pm 0.3 \text{ nm}$, which agrees with previous studies [52,53]. For the magnetic dead-layer thickness $t_0 = 3.85 \pm 0.10 \text{ nm}$, the larger value compared to the typical value of 1.4 nm can be attributed to laser heating and interfacial lattice mismatch [33]. This result is also consistent with optical measurements (see more details in S4) [33]. These fitting results further validate the robustness of our model. For Ni/ CuO_x orbitronics THz emitter, we adopted the product of conductivity and thickness, $\sigma_{\text{NM}} t_{\text{NM}} = 2.84 \times 10^{-3} \Omega^{-1}$, according to resistance measurements (see more details in S4) [33]. Based on the theoretical simulations, we determined the magnetic dead-layer thickness $t_0 = 3.85 \pm 0.10 \text{ nm}$. The deduced orbital diffusion length in the Ni layer is $\lambda_L^{\text{Ni}} = 15.0 \pm 2.8 \text{ nm}$, which is larger than the spin-diffusion length by a factor of 4.5 for the Ni/Pt system (see more details in S4) [33,54].

In the Ni/NM heterostructure, the spin current induced by the ultrafast laser pulse in Ni is converted to orbital current during the transport process, and then both the spin- and orbital currents are injected into the NM layer. When NM = Pt, the spin current is converted to transient charge currents by ISHE in Pt, generating THz radiation. In the case of NM = CuO_x , OAM is converted to transient charge currents by IOREE at the Ni/ CuO_x interface, generating THz emission. Increasing t_{Ni} thickness enhances the orbital accumulation at the Ni/ CuO_x interface, which explains the increase in THz signals with t_{Ni} . The long tail for thick t_{Ni} can be ascribed to the shunting effect and the laser absorption of the Ni layer [55]. Compared with the typical spintronic THz generator Ni/Pt, long-range orbital propagation results in the THz amplitude of the Ni/ CuO_x system peaking at the thicker Ni layer.

Additionally, the magnetic dipole radiation is calculated by combining the THz signals from both substrate-side and film-side pumping. However, the magnetic dipole radiation

exhibits a negligible impact on the thickness-dependence experimental results and, therefore, is not considered further in this study (see Supplemental Material, S7 for more details) [33].

For the $\text{Al}_2\text{O}_3/\text{Ni}/\text{CuO}_x$ structure, the top and bottom interfaces of Ni are asymmetric, which could lead to various self S-C effects in Ni, such as the inverse spin Hall effect (ISHE) and the anomalous Hall effect (AHE). We systematically investigated the THz emission from the $\text{Al}_2\text{O}_3/\text{Ni}(9)/\text{CuO}_x(t_{\text{CuO}_x})$ samples, where the thickness of the CuO_x layer t_{CuO_x} was set to 3, 5, 7, and 11 nm (see Supplemental Material, S6 for more details) [33]. Note that the $\text{Al}_2\text{O}_3/\text{Ni}/\text{CuO}_x$ samples were obtained by exposing the $\text{Al}_2\text{O}_3/\text{Ni}/\text{Cu}$ samples to the atmosphere at room temperature for 48 h to natural oxidization. The asymmetry of $\text{Al}_2\text{O}_3/\text{Ni}$ and Ni/CuO_x interfaces is the same for different samples. Since the self S-C effects are closely related to the two interfaces, the THz signals originating from self S-C effects in Ni will be independent of the CuO_x layer thickness. However, the amplitude of the THz signals decreases as the CuO_x layer thickness increases, which is not consistent with the feature of the self-induced ISHE THz emission. More importantly, a marked time delay with an increase of CuO_x thickness is observed, a phenomenon that cannot be explained by either ultrafast demagnetization or self S-C effects. As well established, this distinguished observation of the delay in the THz peak is a fingerprint of ballistic orbital transport [20,56]. Therefore, we found conclusive evidence that the THz emission from Ni/CuO_x samples mainly stems from the IOREE mechanism.

In summary, we systematically investigated the THz emission from the Ni/CuO_x heterostructures with different layer thicknesses for Ni and demonstrated the IOREE mechanism for THz emission. Considerable laser-excited orbitronic THz

radiation was achieved, with an intensity close to 20% that of the Ni/Pt spintronic THz emitter. We found a polarity reversal of the THz waveform by reversing the external magnetic field, and the THz intensity of Ni/CuO_x can be effectively tuned by the oxidation level of CuO_x . We built up a theoretical model that includes both spintronic and orbitronic contributions, and the dependence of the THz amplitude for Ni/CuO_x and Ni/Pt on the thickness of the Ni layer could be described well by the corresponding theoretical model. Further analysis indicates that the orbital diffusion length $\lambda_L^{\text{Ni}} = 15.0 \pm 2.8$ nm is much larger than the spin-diffusion length $\lambda_S^{\text{Ni}} = 3.3 \pm 0.3$ nm in the Ni layer, resulting in the THz amplitude of the Ni/CuO_x system peaking at the thicker Ni layer. This work not only sheds light on the efficient generation of THz emission based on ultrafast orbit-to-charge conversion by the IOREE, but also provides guidance for the design and fabrication of orbitronic THz emitters.

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