

Terahertz harmonic generation across the Mott insulator-metal transition

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High harmonic generation (HHG) in the terahertz (THz) regime is an emerging field that has already provided new insights into fundamental low-energy processes in Dirac materials and high- T_C superconductors. Here, we demonstrate THz harmonic generation across the Mott insulator-metal transition in rare-earth nickelates ($RNiO_3$, R = rare-earth atom). The THz harmonic signal shows distinct characteristics in the three different accessible phases: the intensity of harmonics increases upon cooling in both the low-temperature antiferromagnetic (AFM) insulating and high-temperature paramagnetic (PM) metallic phases, while this trend is reversed in the intermediate PM insulating phase. Using single- and two-band Hubbard models, we find different dominant origins of the THz harmonics in different phases: strong spin-charge and orbital-charge coupling in the AFM insulating phase, intraband currents from renormalized quasiparticles with temperature-dependent scattering rate in the PM metallic phase, and the reduction of the charge carrier density due to the opening of the Mott gap in the PM insulating phase. These results and mechanisms significantly differ from those observed upon excitation in the optical regime. Our study lays the foundation for THz HHG physics in Mott and other strongly correlated systems and offers design principles for efficient THz HHG from these systems.

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I. INTRODUCTION

A fundamental manifestation of nonlinear light-matter interaction is high harmonic generation (HHG)—the emission of light at integer multiples of the driving frequency. Since the advent of intense femtosecond lasers, the number of HHG-based studies on different material systems is continuously growing, starting from atomic and molecular gases to liquids and complex solids [1–4]. In parallel, significant progress has also been made in theoretical modeling of HHG from different classes of materials, including solids [5,6], strongly correlated systems [7–12], quantum spins [13], spin-orbit coupled systems [14], and altermagnets [15]. HHG spectra have been extensively used as a probe to investigate the electronic structure of atoms and molecules, the band structure of solids, and interatomic bonding [16–20]. Furthermore, HHG is the primary method to generate attosecond pulses of frequencies ranging from the extreme ultraviolet to soft x-rays

[21,22]. However, these activities have so far been primarily carried out using optical (near-infrared to near-ultraviolet) drive. Similar activities using a terahertz (THz) drive started only recently after table-top and electron accelerator-based THz sources with sufficient intensity have become available [23,24]. Further, the theoretical description of the THz HHG from different material systems is not yet fully developed. Nevertheless, within a short span of time, studies based on THz HHG have shown promise in diverse applications, e.g., efficient THz harmonic emission from Dirac and topological materials [25–31], identifying dynamics of Higgs and charge-density wave modes in superconductors [23,32–36], light-wave electronics of the surface states in Dirac materials [37], light field-driven ultrafast spin current generation [38], etc. Recently, additional mechanisms have also been proposed for THz harmonic generation, based on the electrical polarization, spin current, and magnetization [39–41].

THz studies can play a crucial role in exploring strongly correlated materials. The macroscopic phases of these materials result from interactions among fundamental charge, spin, orbital, and lattice degrees of freedom [42]. Thus, understanding the structure and dynamics of these interactions is central to advancing the fundamental physics of such materials and exploiting their properties for technological applications. Since the energy scale of these interactions often overlaps with the THz band (~ 0.1 – 10 THz) [43], one can directly probe their role in the nonlinear responses of these materials

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by performing THz HHG spectroscopy. This can contribute significantly to three broad objectives, namely, developing physics of THz HHG across different strongly correlated materials, using HHG spectra to unravel the complex physics of macroscopic phases and developing strategies to enhance the nonlinear responses desirable for technological applications. Here, we present a THz third harmonic generation (THG) study of one such material system—Mott insulators. Our findings show that the interactions among fundamental degrees of freedom indeed play a dominant role in the nonlinear response of Mott insulators to the THz driving field.

Prior to our study, there exist a few HHG studies on Mott insulators but only with optical excitations [44–49]. A recent HHG study on the Mott insulator Ca_2RuO_4 using midinfrared (MIR) excitation revealed an anomalous temperature dependence of the HHG intensity: the intensities of higher-order harmonics, whose photon energies are comparable to or greater than the Mott gap, increase upon lowering the temperature, despite a reduction in the free charge carrier density [49]. This HHG enhancement was explained on the basis of spin-charge coupling present in the antiferromagnetic (AFM) insulating state [48]. The same effect was also predicted in the presence of orbital-charge coupling in a paramagnetic (PM) state. These observations raise several interesting questions: (i) Can Mott insulators also emit high harmonics upon THz excitation, considering that the THz photon energy (a few meV) is three orders of magnitude smaller than the Mott gap (typically of the order of 1 eV)? (ii) If yes, what is the mechanism underlying HHG in the THz regime? (iii) Do higher-order THz harmonics, similar to that observed upon MIR excitation, show anomalous temperature dependence? Even if they do show an anomalous temperature dependence, we may not observe it experimentally because then we need to detect the order of 100th harmonics (to match the energy scale of the Mott gap) and the fact that the intensity of the harmonics usually decreases with increasing order. (iv) What is the HHG response in the vicinity of the Mott insulator-metal transition (IMT) in the correlated metallic state? Above the transition temperature (T_{IM}), HHG in Ca_2RuO_4 has not been explored. In a similar correlated system, V_2O_3 , HHG was not observed above T_{IM} upon optical excitation [50]. However, THz HHG was recently observed in the quantum critical metal CaRuO_3 [51] and in elemental transition metals with nonzero spin-Hall conductivity [52].

To explore the possibility of THz harmonic emission from Mott insulators and to address the abovementioned questions, we performed THz THG experiments on rare-earth nickelates (RNiO_3 , R = rare-earth atom)—a prototypical Mott insulator well known for exhibiting an IMT [53,54]. The typical temperature-dependent phase diagram of nickelates consists of three different regimes: a low temperature AFM insulating regime, an intermediate temperature PM insulating regime and a high temperature PM metallic regime. Not only did the nickelates exhibit a clear THz THG signal in all three regimes, but they also showed distinctly different responses in the different regimes of the phase diagram. Remarkably, the THG also exhibits an anomalous temperature-dependence, i.e., strong THG enhancement upon lowering the temperature in the AFM-insulating regime. Our simulations of the experimental results using single and multiband Hubbard models

reproduce the characteristic features of the THz THG from Mott insulators. Our results demonstrate that (i) the THz HHG from Mott insulators is indeed possible, and (ii) in contrast to HHG upon optical excitation, the THz HHG enhancement in the AFM insulating phase occurs for all orders of harmonics. This shows that the THz HHG does not require photon energies comparable to the width of the Mott gap. The strong sensitivity of the THz HHG spectra on low-energy many-body interactions furthermore suggests that they can probe fundamental processes in complex systems and provide guidance for future technological applications.

II. EXPERIMENTAL DETAILS

Two high quality films of LaNiO_3 and NdNiO_3 were fabricated on LaAlO_3 (100) and LSAT (100) single crystal substrates, respectively, using pulsed laser deposition technique. To confirm the phase purity and high quality of the films, structural characterizations of the films were performed on a Panalytical x-ray diffractometer. The Mott IMT and PM to AFM transition and their respective transition temperatures were determined by performing DC resistivity measurements. The resistivity measurements were performed using the four-probe method in the temperature range of 2–300 K on a Quantum Design-made physical property measurement system (PPMS). Finally, the THz THG measurements on these films were performed using intense narrowband multicycle THz pulses ($E_{\text{THz}} \sim 100$ kV/cm) with central frequency $\omega = 0.3$ THz. The THG signal from the transmitted THz radiation from the films was filtered using two bandpass filters with central frequency $3\omega = 0.9$ THz, which significantly suppress the residual radiation at the fundamental frequency. The THG signal was then detected in the time domain using a 2 mm thick ZnTe crystal. For further details on the thin film growth, structural and electrical characterizations, and THz THG measurements, see the Supplemental Material (SM) [55].

III. RESULTS AND DISCUSSION

Figure 1(a) shows the temperature-dependent DC resistivity of $\text{LaNiO}_3/\text{LAO}$ (100) and $\text{NdNiO}_3/\text{LSAT}$ (100) films. It is clear that the $\text{LaNiO}_3/\text{LAO}$ (100) film remains a PM metal down to low temperature, while the $\text{NdNiO}_3/\text{LSAT}$ (100) film exhibits a clear first-order IMT at ~ 150 K. The $\text{NdNiO}_3/\text{LSAT}$ (100) film also exhibits an AFM to PM transition at ~ 106 K, as inferred from the inverse temperature derivative of its resistivity [see inset of Fig. 1(a)]. Both films, particularly at low temperatures, emit a strong THG signal upon excitation with 0.3 THz pulses. An exemplary THG waveform emitted from the LaNiO_3 film is shown in Fig. 1(b) and its Fourier transform is plotted in Fig. 1(c). The observation of a strong intensity peak at 0.9 THz confirms the THG emission from the LaNiO_3 film.

Figure 2(a) shows the temperature-dependent normalized THG intensity ($I_{3\omega}$) of the PM metallic LaNiO_3 film. The THG intensity increases monotonously upon lowering the temperature down to the lowest measured temperature of $T \sim 20$ K. The same holds true for the PM metallic state of the NdNiO_3 film [see Fig. 2(b)]. However, below T_{IM} , the THG intensity starts decreasing and this trend continues down to

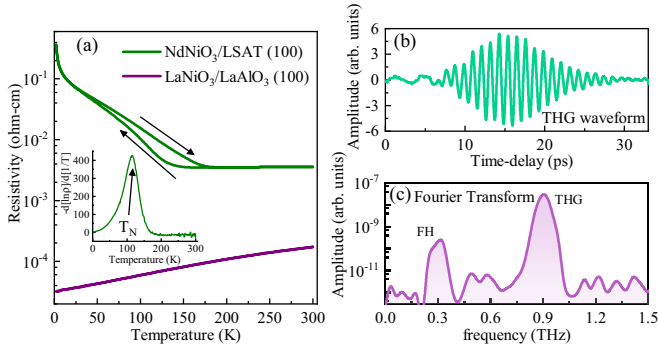


FIG. 1. (a) Temperature-dependent resistivity of LaNiO_3 and NdNiO_3 films. The arrows show the heating and cooling cycles. The inset shows T_N of the NdNiO_3 film in the cooling protocol determined by the peak in $-\frac{d[\ln \rho]}{d[1/T]}$. (b) THG temporal waveform measured after two 0.9 THz bandpass filters for the LaNiO_3 film at $T = 22$ K. Its Fourier transform is shown in (c). The peaks at 0.3 THz and 0.9 THz represent the driving pulse and THG signal, respectively.

the magnetic transition temperature (T_N). Remarkably, below T_N , the THG intensity starts rising sharply and monotonously upon cooling down to the lowest temperature, showing an increase in the THG intensity by more than one order of magnitude at ~ 5 K compared to that at T_N . Such an enhancement in the THG intensity below T_N could occur due to the increased transmission of the THz radiation in the highly insulating state of the film. However, as shown in Sec. S5 in the SM [55], the increased transmission of the THz radiation has negligible effect on the observed temperature dependence of the THG intensity.

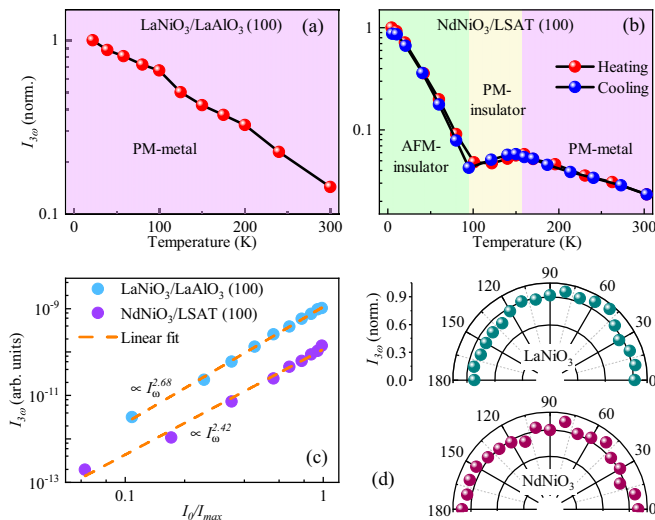


FIG. 2. Temperature-dependent THG intensity for (a) LaNiO_3 and (b) NdNiO_3 films. Different colored regions in (b) represent the temperature-dependent different electronic states of the NdNiO_3 film. (c) THG intensity as a function of the fundamental THz intensity for LaNiO_3 ($T = 22$ K) and NdNiO_3 ($T = 10$ K) films, plotted on a log-log scale. Dotted lines are linear fits to the experimental data. (d) Polar plots of THG from LaNiO_3 and NdNiO_3 films at 10 K as a function of the driving THz pulse polarization angle.

We plot the THG intensity ($I_{3\omega}$) as a function of fundamental THz intensity (I_ω) for both the films in Fig. 2(c). Ideally, one expects a cubic relation $I_{3\omega} \propto I_\omega^3$; however, for both films, the exponent is significantly smaller than 3. This indicates that the THG emission from the nickelate films originates from light-matter interaction in the nonperturbative regime [27]. Typically, the nonperturbative light-matter interactions occur when the driving field is quite strong and/or for higher-order harmonics (e.g., seventh and higher-order harmonics). However, in complex materials such as Mott insulators, topological insulators, and high- T_C superconductors, the nonperturbative light-matter interactions can occur even with moderate driving field and for the lower-order harmonics [26,34,49]. Further, one might expect a crystal-orientation-dependent THG response due to strain-induced modifications in the magnitude of the O-2p and Ni-3d orbital polarizations in different directions [56]. To determine the directional dependence of the THG, THz polarization-dependent THG measurements were performed at $T = 10$ K for both films. As shown in Fig. 2(d), the THG intensity in both cases is nearly independent of the THz polarization orientation, which indicates that the sum of the contributions to the THG signal is isotropic. This is consistent with the fact that the films are isotropically strained due to the cubic structure of the underlying substrates. Consequently, the magnitude of the orbital polarization should be identical along the two orthogonal in-plane directions.

The existing theoretical models for Dirac materials and superconductors can successfully reproduce their experimental THz HHG results and thus have significantly contributed to the understanding of the THz HHG mechanisms in these systems [23,25–28,32–36]. However, such theoretical developments for Mott insulators have been limited to HHG in the optical regime only [44–48]. Optical HHG from Mott insulators can be modeled using the nonequilibrium DMFT formalism [44,48,57]. HHG in single-band Mott insulators originates both from the intra-Hubbard-band dynamics of doublons (doubly occupied states) and holons (empty states) and from their recombination [48]. A similar situation can be expected in two-orbital systems in the PM state. Here, the hopping between singlons (singly occupied states) and triplons (triply occupied states), however, leads to orbital-charge coupling due to the orbital moments of the triplons and singlons [48]. Extending such studies to the THz HHG is challenging for several reasons. First, we must choose a sufficiently low excitation frequency in the simulation in order to qualitatively reproduce the THz HHG experiments, where the photon energy is three orders of magnitude smaller than the Mott gap. This requires simulations up to long times, which is numerically challenging (see Sec. S6 [55]). Second, switching from the optical to the THz regime also significantly changes the dynamics of the excited charge carriers. For example, carrier production in the optical regime occurs via multiphoton absorption, while in the THz regime it occurs from tunneling across the bandgap [58–60]. Also, in the THz regime, the contribution to HHG from intraband charge acceleration is expected to exceed that from interband tunneling [61]. Finally, in the AFM phase of the nickelates, spin-charge and orbital-charge couplings are simultaneously active, and their interplay needs to be considered in the simulation.

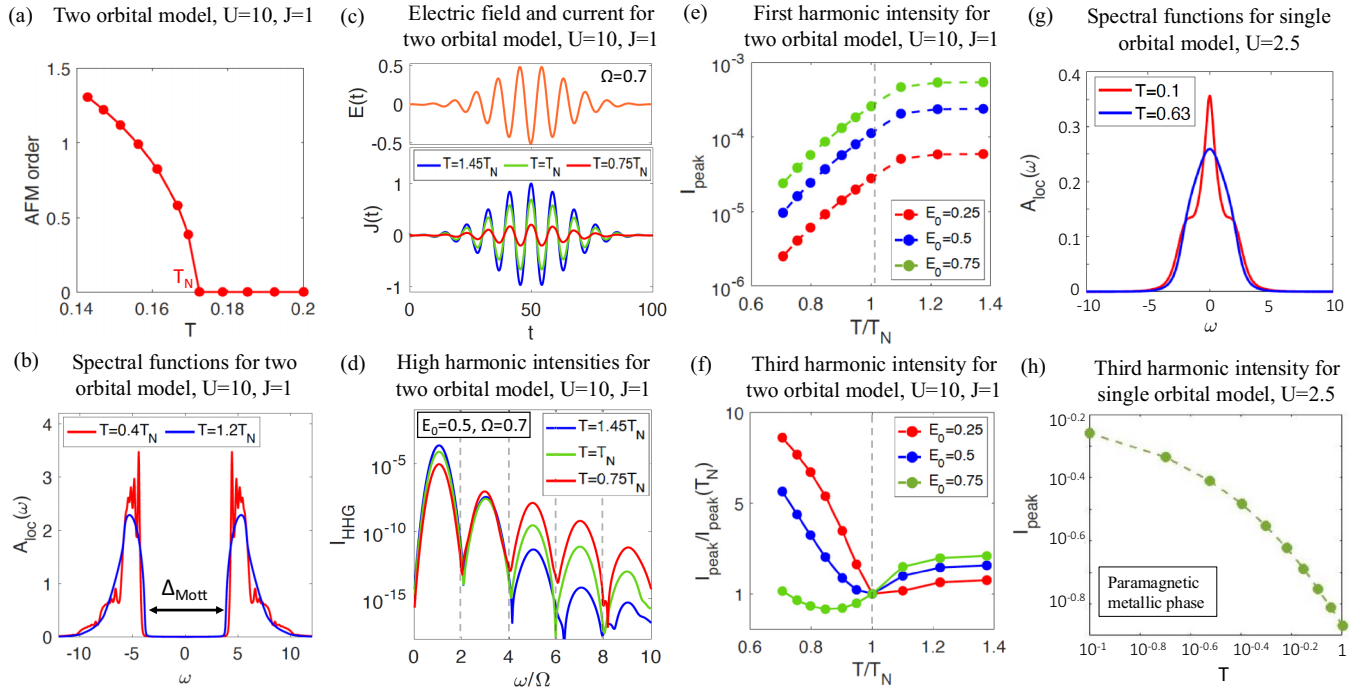


FIG. 3. DMFT results for the two-orbital repulsive Hubbard model with $U = 10$, $J = 1$. (a) AFM order as a function of temperature with the Néel temperature T_N indicated. (b) Single-particle local spectral function $A_{\text{loc}}(\omega)$ below and above T_N . (c) Electric field pulse $E(t)$ with $\Omega = 0.7$ (upper panel) and the resultant current $\mathcal{J}(t)$ (not to scale) for different temperatures (lower panel). (d) High harmonic intensities for $E_0 = 0.5$ and $\Omega = 0.7$ for different temperatures [obtained from the Fourier transform of the $\mathcal{J}(t)$ shown in (c)]. (e) Peak values I_{peak} of the first harmonic intensity and (f) I_{peak} of the third harmonic intensity as a function of temperature for different electric field strengths E_0 . (g) Local spectral function $A_{\text{loc}}(\omega)$ of the single-orbital repulsive Hubbard model with $U = 2.5$ for different temperatures. (h) Peak values I_{peak} of the third harmonic intensity as a function of temperature for the single-orbital Hubbard model with $U = 2.5$.

To account for all these relevant factors, we use the two-orbital Hubbard model with Hamiltonian

$$\begin{aligned}
 H = & -t_{\text{hop}} \sum_{\langle ij \rangle, \sigma} \sum_{\alpha=1,2} c_{i,\alpha\sigma}^\dagger c_{j,\alpha\sigma} + U \sum_i \sum_{\alpha=1,2} n_{i,\alpha\uparrow} n_{i,\alpha\downarrow} \\
 & - \mu \sum_i \sum_{\alpha=1,2} (n_{i,\alpha\uparrow} + n_{i,\alpha\downarrow}) + (U - 2J) \sum_{i,\sigma} n_{i,1\sigma} n_{i,2\sigma} \\
 & + (U - 3J) \sum_{i,\sigma} n_{i,1\sigma} n_{i,2\sigma}, \quad (1)
 \end{aligned}$$

where α and σ denote the orbital and spin indices, respectively, t_{hop} is the nearest-neighbor hopping amplitude between sites i and j , U is the intraorbital Hubbard repulsion, J the Hund coupling, and μ the chemical potential (for further details, see Sec. S6 [55] and Refs. [62–66] therein). We use a code based on the NESSi library [57] and a noncrossing approximation (NCA) impurity solver [67,68] to analyze the dynamics of large-gap Mott insulators driven by an electric field pulse $E(t) = E_0 f_g(t) \sin(\Omega t)$, where Ω is the pulse frequency and f_g is a Gaussian envelope function [Fig. 3(c)]. In such a driven system, charge carriers are created with a density controlled by the electric field strength E_0 , and their kinematics are coupled to the correlated background of spin and orbital moments.

In a single-orbital system, strong spin-charge coupling of the charge excitations (doublon-holon pairs) with the AFM spin background gives rise to an increase in HHG below the T_N [48]. With decreasing temperature, the coherence between

the doublons and holons increases because of suppressed thermal fluctuations in the spin background. This reduces the phase cancellations in the emitted light, enhances the high harmonic intensities, and leads to a more pronounced HHG plateau [44,46,48].

In a two-orbital system, although the basic concept of coupling of the charge excitations to a correlated background remains the same, there are some differences because the background now features both spin and orbital moments. For Mott insulators with positive Hund coupling ($J > 0$) the anomalous temperature dependence of the HHG peaks is observed for the fifth and higher harmonics even above the Néel temperature, because the hopping of singlons (singly occupied sites) and triplons (triply occupied sites) in a background of high-spin doublons can leave behind low-spin configurations. If we decrease the temperature further below the Néel temperature (T_N), spin-charge coupling with excitation energies of order $J_{\text{ex}} \ll J$ is activated, and we observe an increase in high harmonic intensities with decreasing temperature for the third and higher harmonics. In Fig. 3(a) we show the AFM order as a function of temperature, while Fig. 3(b) shows the equilibrium single-particle spectral function at different temperatures for the two-orbital Hubbard model with $U = 10$, $J = 1$, which are approximately the realistic parameters for nickelates [69]. Below T_N , the Mott gap splits into sharp subbands due to the formation of spin-polarons. We measure the high harmonic intensity $I_{\text{HHG}}(\omega)$ from the Fourier transform of the current $\mathcal{J}(t)$ as $I_{\text{HHG}}(\omega) = |\omega \mathcal{J}(\omega)|^2$. $\mathcal{J}(t)$ is

shown for a few temperatures in Fig. 3(c), while Fig. 3(d) plots $I_{\text{HHG}}(\omega)$ for the two-orbital Hubbard model with $U = 10$, $J = 1$ and driving frequency $\Omega = 0.7$ for different temperatures. Due to inversion symmetry, we get peaks at odd multiples of the driving frequency Ω . The increase in the intensity of the higher harmonics with decreasing temperature (both above and below T_N) is clearly seen.

A different behavior is found for the first and third harmonics. In Figs. 3(e) and 3(f), we plot the peak values of the first and third harmonic intensities, respectively, as a function of temperature and at different fixed electric field strengths. We start with $E_0 = 0.25$ (red curve) and a temperature above T_N in the PM insulating region. As we gradually decrease the temperature, we observe a decrease in the first and third harmonic intensities. This is due to the increase of the Mott gap with decreasing temperature and the suppression of the density of charge carriers. Once we are below T_N , we observe an increase in the THG intensity with decreasing temperature. In this region, although the number of the induced charge carriers still decreases with decreasing temperature (as reflected in the decreasing first harmonic intensity), there is a strong spin-charge coupling of the charge carriers with the ordered background. The formation of spin polarons is evident from the appearance of sharp peaks in the single-particle spectral functions at equilibrium below T_N [see Fig. 3(b)]. Next, we increase the electric field strength to $E_0 = 0.5$ and observe an increase in the overall first harmonic intensity due to the increased density of charge carriers. At the same time, the rate of increase of the THG intensity decreases. If we increase the electric field strength further to $E_0 = 0.75$, the THG intensity decreases even below the Néel temperature but starts increasing below about $0.8T_N$ [Fig. 3(f)]. A higher electric field leads to a stronger disordering of the spin background and we need stronger correlations in the initial state to observe the increase in THG intensity with decreasing temperature, which is achieved at $T \ll T_N$. In this context, it is interesting to note that the experiments in Ref. [49], which do not observe any THG increase, used a field strength in the MIR range which is more than two orders of magnitude higher than that in the present study.

In the high-temperature PM metallic phase, the physics can be approximately described by a single-orbital model. To analyze the THG in the metallic region, we use the single-orbital Hubbard model with $U = 2.5$ and a one-crossing approximation (OCA) impurity solver [68,70], which is computationally more costly than NCA, but provides a better description of metallic states. The local spectral function of this model is shown in Fig. 3(g). At high temperatures, the metallic spectrum is relatively broad, but at lower temperatures, a sharp low-energy peak emerges, indicative of coherent quasiparticle excitations. Consistent with this, we observe that the THG intensity increases with decreasing temperature in the metallic phase, as seen in Fig. 3(h). The THG in the metallic phase can be attributed to the intraband dynamics of the quasiparticles, which for large field amplitudes results in a nonlinear response [51]. Due to strong electronic correlations, nickelates often exhibit metallic states with both incoherent and coherent features, indicative of energy (or frequency and temperature) dependent scattering rates [71,72]. Below T_{IM} , the Mott gap opens and increases with decreasing temperature, leading to

a suppression of the induced charge carrier density. At sufficiently high temperatures, this effect dominates over the orbital-charge coupling effect on THG, leading to a decrease in THG intensity with decreasing temperature in the PM-insulating regime. In addition, the coexistence of insulating and metallic domains in the temperature range from ~ 60 K to ~ 160 K for the $\text{NdNiO}_3/\text{LSAT}$ (100) film [Fig. 1(a)] can increase the charge carrier scattering due to the increased phase inhomogeneity [73,74], leading to a further reduction of the THG intensity below T_{IM} .

IV. CONCLUSIONS

In summary, we experimentally demonstrated THz THG from a prototypical Mott insulator and successfully modeled the observed temperature dependence of the THG on the fundamental quasiparticle interactions. The THz THG is visible in all the three different regimes of the nickelate phase diagram along with a remarkably sharp enhancement in the AFM insulating regime. This enhancement occurs due to the combined effect of spin-charge and orbital-charge couplings. In the PM correlated metallic state, the THG mechanism is dominated by the responses of heavy quasiparticles near the Fermi level and their temperature-dependent scattering rate. In the intermediate PM insulating state, the characteristic temperature dependence of THG is governed by the size of the Mott gap.

The observation of the THz THG intensity enhancement in the AFM insulating regime breaks the condition recently set for optical HHG that the photon energy for a given harmonics should exceed the Mott gap for its intensity enhancement. Its temperature dependence is reproducible only by considering more realistic scenario, i.e., the multiorbital nature of the Mott insulators and the interplay of both spin-charge and orbital-charge couplings at the microscopic level. This suggests that the THz HHG spectra of strongly correlated materials and their underlying mechanisms are more sensitive to microscopic details than the optical HHG counterparts. Thus, our findings may inspire and contribute to further such studies, for example: (i) Exploring THz HHG physics in other strongly correlated systems, particularly in $3d$ to $5d$ transition metal oxides [75], where the role of other factors such as spin-orbit coupling, topology, and crystal fields in the HHG can be explored. (ii) Extending the range of efficient THz HHG materials beyond Dirac and topological materials. Favorable conditions for efficient HHG can be achieved in multiple ways, including chemical doping, photodoping, or the fabrication of thin films and heterostructures. (iii) Using the strong sensitivity of the THz HHG to low-energy interactions to gain insights into the microscopic physics of complex systems.

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