

Signatures of Kondo-lattice behavior in the two-dimensional ferromagnet Fe_3GeTe_2

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Fe_3GeTe_2 is a paradigmatic van der Waals magnet with the full microscopic picture of magnetic interactions still under debate. Here, we use scanning tunneling microscopy and spectroscopy at 1.1 K to map out the low-energy physics on the surface. In agreement with previous works, we observe a spatially varying Fano line shape that has been ascribed to a Kondo resonance. In addition, we identify a small gap of ≈ 2.3 meV width on top of the larger-energy Fano resonance. We suggest that this gap is the signature of a coherent Kondo lattice that has been proposed to originate from hybridization of a strongly dispersing and a flat band both derived from d electrons of the Fe atoms in the lattice. Adding magnetic adatoms on the surface hardly influences the magnetic signatures of the substrate, indicating the robustness of the intralayer Kondo correlations against magnetic adsorbates.

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I. INTRODUCTION

The interplay between localized magnetic moments and itinerant conduction electrons in strongly correlated materials has long been a subject of intense research [1]. One fascinating manifestation of this interaction is the Kondo lattice, where the coupling between localized spins and conduction electrons leads to a complex interplay between Kondo screening and magnetic ordering [2–4]. Kondo-lattice systems have been extensively studied in bulk intermetallic compounds containing rare earth atoms. The localized magnetic moments are provided by f electrons that are exchange coupled to the itinerant conduction electrons, leading to Kondo screening and the formation of a Kondo lattice [5–7]. In contrast, in d electron systems, heavy-fermion behavior is typically not observed owing to the absence of strongly localized magnetic moments. However, hybrid systems, in which localized electrons originate from an adsorbate structure containing transition metal atoms, coupled to an electron bath in the substrate, have exhibited signatures of extended Kondo correlations [8–10].

With the advent of two-dimensional (2D) materials, new opportunities for exploring competing interactions and correlation effects entered the stage. In reduced dimension, quantum confinement and enhanced electronic correlations add versatility and novel phenomenology, including superconductivity, Mott insulating behavior, charge-density

modulations [11], and in particular also ferromagnetism [12] and Kondo-lattice behavior [13].

It was only in the recent years that the first 2D magnets were isolated. One of the first was CrI_3 , which shows ferromagnetic or antiferromagnetic behavior depending on the number of layers [14], followed by ferromagnetic $\text{Cr}_2\text{Ge}_2\text{Te}_6$, which loses its magnetic order in the monolayer limit [15]. Fe_3GeTe_2 is a ferromagnet with a high T_C of about 220 K [16–18] even at monolayer thickness [19], which makes it particularly attractive for spintronic applications. However, closer examination of this material reveals that its magnetic behavior is far more complex than that of a simple ferromagnet. A large set of experimental observations revealed seemingly contradictory results. Signatures of both itinerant ferromagnetism and large effective electron mass indicating the presence of localized magnetic moments were observed, launching also intense theoretical efforts to resolve these apparent contradictions [20–23]. The coexistence of itinerant and local magnetism has been explained by either the multi-orbital character of the Fe atoms [24] or two distinct Fe sites exhibiting different Mott and Hund correlation strengths [23,25–28].

The role of orbital- and site-differentiated magnetic contributions are also central to the debate about heavy-fermion behavior and Kondo physics in this system. In particular, the observation of a Kondo lattice would otherwise be incompatible with a ferromagnetic ground state. However, despite indications of Kondo physics, an unambiguous signature of a Kondo lattice remains elusive. Scanning tunneling microscopy (STM) experiments revealed a Fano-shaped dip at the Fermi level, whose line profile varies across the surface. This spectral fingerprint was assigned to a Kondo resonance. Given the omnipresent Fano resonance (albeit with variations in shape) across the surface combined with complementary indications of heavy-fermion behavior, it was suggested that

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the Kondo impurities form indeed a Kondo lattice [13,22]. However, this interpretation was challenged by more recent STM experiments, which suggested that the observed features in the same energy range may instead arise from phonon-magnon excitations [29] or from the suppression of the density of states by disorder in the correlated system [30]. This discussion echoes the earlier apparent controversies in the bulk material, where two-site and multiorbital models were introduced to capture the contradicting observations. A crucial step toward resolving the Kondo-lattice interpretation would be the detection of a hybridization gap superimposed on the Fano line shape as a clear signature of Kondo-lattice formation [5,7,31].

In this work, we contribute to the ongoing debate by presenting high-resolution scanning tunneling microscopy and spectroscopy measurements of low-energy excitations on the surface of a bulk Fe_3GeTe_2 (purchased from HQ graphene and cleaved in ultrahigh vacuum) sample at 1.1 K. Our spectra reproduce the previously observed Fano line shape while also revealing an additional small gap of ≈ 2.3 meV width superimposed on the broader Fano feature. This small gap rapidly disappears with increasing temperature, whereas the overall Fano line shape remains intact. We propose that this gap represents the characteristic hybridization gap, providing further indication for the Kondo-lattice and heavy-fermion behavior in Fe_3GeTe_2 . Additionally, we employ magnetic adatoms as probes to assess the balance of competing interactions in the low-temperature many-body state. Our results show that individual iron (Fe) and manganese (Mn) adatoms mainly affect the asymmetry parameter of the Fano line shape owing to additional cotunneling paths but do not alter the other Kondo parameters. This indicates that the many-body phase coherence in the substrate remains robust against small magnetic perturbations.

II. RESULTS

A. Topography and electronic structure

The crystal structure of Fe_3GeTe_2 [shown in Fig. 1(a)] consists of Te-terminated van-der-Waals-stacked layers. The Fe atoms are located in two different Wyckhoff positions. The central plane is formed by Fe-Ge, with the Fe atoms typically referred to as Fe2, sandwiched between planes of hexagonal Fe arrangements, where the Fe atoms are called Fe1. The Fe atoms not only differ in their crystal-field symmetries, but also in their contribution to the electronic structure and magnetic properties. Indeed, the seemingly contradictory magnetic behavior resembling Stoner-type and Heisenberg-type magnetism has been discussed to be rooted in the different types of Fe atoms, likely arising from different Hund coupling strengths [20].

Large-scale STM images taken after cleaving the bulk crystal in ultrahigh vacuum reveal atomically flat terraces of the terminating Te layer [Fig. 1(b)]. Alongside the atomic corrugation, a nonperiodic modulation of the apparent height is observed. A magnified view in the inset of Fig. 1(b) highlights one of such depressions, approximately 60 pm in apparent depth. These features have been attributed to randomly distributed Fe2 vacancies in the near-surface layers, which

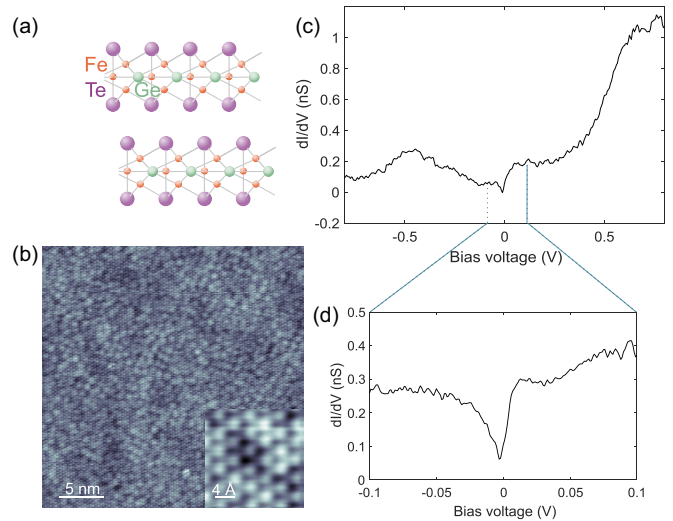


FIG. 1. Overview of Fe_3GeTe_2 . (a) Crystal structure of Fe_3GeTe_2 . The atomic species are color coded as shown. (b) STM topography of the Fe_3GeTe_2 surface. Inset: close-up view on some defects in an atomically resolved STM image ($V_s = 200$ mV, $I_t = 150$ pA). (c) dI/dV spectrum obtained on the Fe_3GeTe_2 surface. ($V_s = 800$ mV, $I_t = 300$ pA, $V_{\text{mod}} = 5$ mV). (d) Close-up view on dI/dV spectrum around the Fermi energy with an asymmetric dip ($V_s = 100$ mV, $I_t = 300$ pA, $V_{\text{mod}} = 1$ mV). Analysis of STM topographs and spectroscopy data was performed with the WSXM [32] and SpectraFox [33] software packages.

locally modify the density of states [22]. Notably, despite these inhomogeneities, long-range ferromagnetism remains intact, persisting over domains spanning several hundred nanometers [21,34,35]. In the following, we examine if this nanoscale disorder influences the Kondo-lattice behavior.

A differential conductance (dI/dV) spectrum [Fig. 1(c)] shows a broad peak centered around -0.5 V and a strong increase in conductance above 0.5 V. Additionally, a narrow dip appears at the Fermi level, as highlighted in the close-up view in Fig. 1(d). The dip features a Fano line shape of about 10 meV width. This signature has been observed previously and was attributed to a Kondo resonance [13,22].

B. Kondo effect

To gain more detailed information, we recorded high-resolution spectra in this energy range at randomly selected locations on the surface. A series of these spectra is presented in Fig. 2(a), all of which exhibit a dip feature around the Fermi level. However, the exact line shape varies across different locations. To quantify these variations and gain insight into the origin of the excitation spectrum, we first fit the data using a Fano function to the conductance $\sigma = A \cdot \frac{(\epsilon + q)^2}{(1 + \epsilon^2)} + \sigma_0$ with $\epsilon = (eV - E_r)/(\Gamma_r/2)$ and a linear background σ_0 . This line shape is expected for tunneling into isolated Kondo impurities on surfaces as well as a Kondo lattice [36,37] and includes the effect of interfering tunneling paths. Here q is the asymmetry parameter which accounts for the ratio between the tunneling paths; E_r is the energy of the resonance and Γ_r corresponds to its full-width. The amplitude of the resonance is accounted by the parameter A . In analyzing the best fits, we notice that

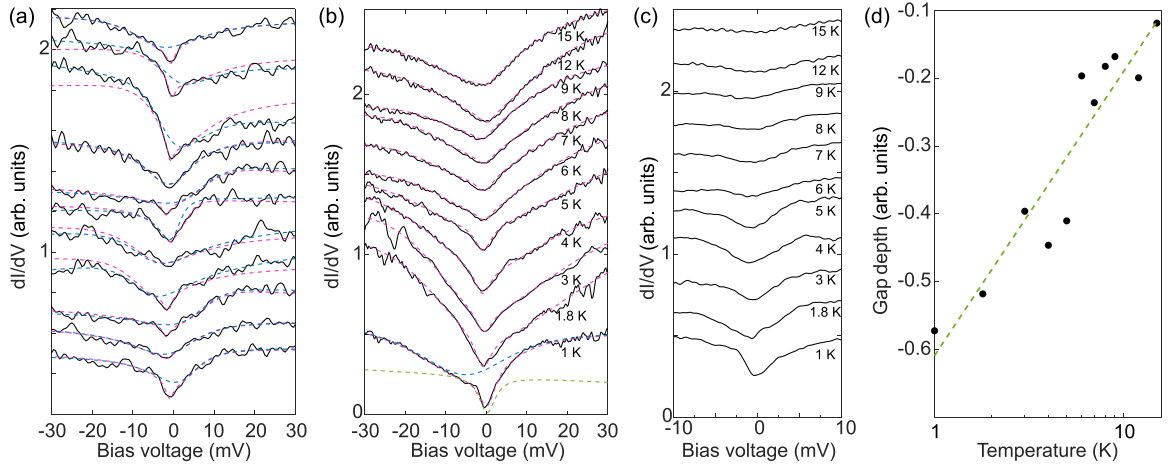


FIG. 2. Low-energy spectra. (a) Normalized dI/dV spectra obtained on multiple locations of the Fe_2GeTe_3 surface. The spectra are fit using two models: one with a single Fano line shape and a linear background, and another combining this with a gap function, as described in the text (solid black: raw data, pink dashed: Fano and gap fit, blue dashed: Fano fit). ($V_S = 40$ mV, $I_t = 300$ pA, $V_{\text{mod}} = 0.5$ mV). (b) Temperature dependence of spectra on the Fe_2GeTe_3 surface ($V_S = 35$ mV, $I_t = 500$ pA, $V_{\text{mod}} = 0.25$ mV). The blue and green dashed lines are the Fano and gap contributions to the dI/dV curve at lowest temperature, respectively. The pink dashed line is the fit to the combined Fano and gap function. (c) Data after subtracting the broad Fano line shape at each temperature. (d) Temperature evolution of the depth of the small gap in logarithmic scale. A logarithmic fit to the data is shown as dashed green line.

the spectra cannot be completely reproduced by a single Fano function. Instead, we find an additional narrow dip at the Fermi level. To fully capture the line shape, we thus add a Lorentzian dip mimicking a not-fully developed gap ($\sigma_{\text{gap}} = A_{\text{gap}} \cdot \frac{\Gamma_{\text{gap}}/2}{(eV - E_{\text{gap}})^2 + (\Gamma_{\text{gap}}/2)^2}$) with A_{gap} the amplitude, Γ_{gap} the gap full-width, and E_{gap} its energy position. Temperature broadening is included as a factor $k_B \cdot T$ in the effective width of both the Fano function and the gap function ($\Gamma = \sqrt{\Gamma_0^2 + (k_B \cdot T)^2}$). This combined function reproduces all experimental data very well [see purple dashed lines in Fig. 2(a)]. The additional gap is of ≈ 2.3 meV width, albeit with site-dependent variations that will be discussed below.

This additional gap has not been reported at a temperature of 4–6 K by Zhang *et al.* [13] or Zhao *et al.* [22]. This discrepancy is easily resolved by raising the measurement temperature. Figure 2(b) shows the dI/dV spectra at a single location measured with increasing temperature. To analyze the presence of the small gap, we again fit a combined function that includes a Fano line shape and an additional gap to the spectra at all temperatures [Fig. 2(b)]. To directly reveal the small gap in the experimental data, we plot in Fig. 2(c) the spectra after subtracting the Fano line shape. The resulting difference spectra clearly highlight the emergence of the small gap at low temperature and its suppression above 5 K. For a more quantitative analysis, we extracted the depth of the small gap from the fit and plot it in Fig. 2(d). It scales logarithmically with temperature. Building on earlier predictions of a Kondo lattice [13,22], supported by the observed temperature dependence [6], we propose that this gap represents the previously elusive evidence of a hybridization gap, thereby supporting the identification of a Kondo lattice. Although most of the experimental and theoretical studies corroborate the Kondo-lattice picture, we note that low-energy features have recently been discussed to originate from other

phenomena. Bansal *et al.* measure at a temperature of 40 mK and observe a sharp gap feature around zero bias that they assign to dynamical Coulomb blockade followed by higher-energy features ascribed to phonon-magnon coupling [29]. Mathimaler *et al.* observed a dip similar to ours in the differential conductance and assigned it to a gap opening due to disorder and correlations captured by the Altshuler-Aronov effect [30]. They also find indications of multifractality in their spatially resolved maps suggesting that the level of disorder competes with complete localization. However, our measurements show no indication of multifractality, as we will show in the next section from spatially resolved spectra.

C. Spatial variations of the Kondo effect

We also investigate the spatial variations of the Kondo resonance and the small dip at the Fermi level. Additionally, we probe their response to single magnetic adatoms that were deposited at 10 K directly into the STM. To do so, we map out the spectral characteristics on the pristine Fe_3GeTe_2 surface and on single Fe [Figs. 3(a)–3(d)] and Mn [Figs. 3(e)–3(h)] atoms by recording dI/dV spectra in a densely spaced grid covering the same area as the topography. We then apply our fit procedure using a Fano line shape combined with an additional dip as described above. We first analyze the characteristics of the Fano contribution. We plot the extracted width Γ_r , asymmetry parameter q , and the position of the resonance E_r in Figs. 3(b)–3(d) for Fe, and Figs. 3(f)–3(h) for Mn.

First, we focus on the pristine background and note that all Kondo parameters exhibit spatial variations. Closer examination of the spatial distribution reveals no clear correlation of the topographic signal with any of the Kondo parameters, i.e., Γ_r , q , or E_r . In calculating the spatially resolved correlation between the topography and the different parameters,

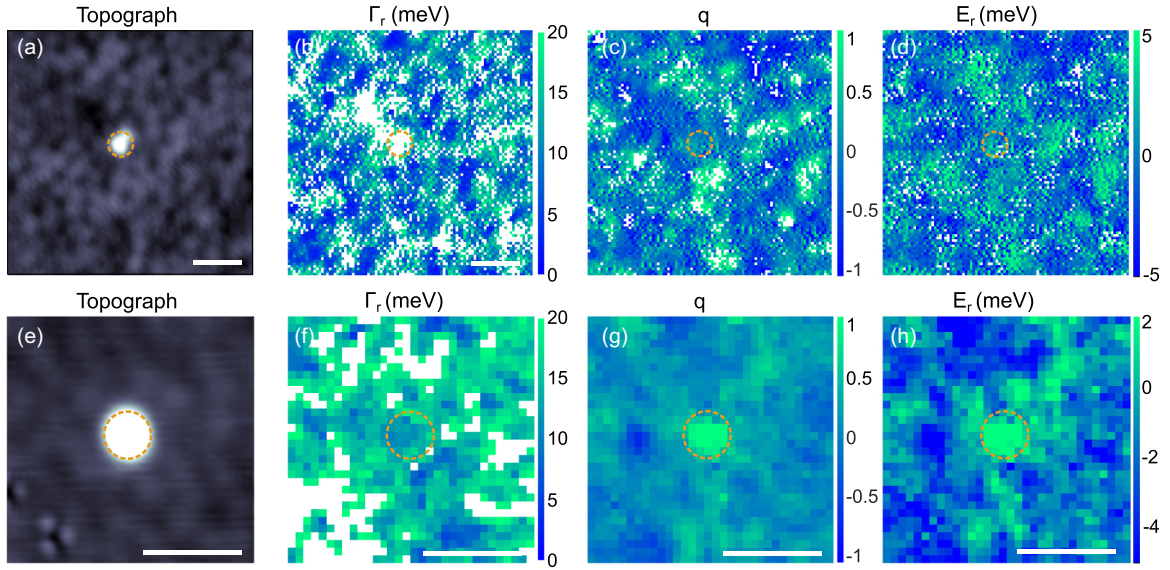


FIG. 3. Spatial variation of Fano parameters in the presence of Fe and Mn atoms. (a), (e) STM topographs around Fe (a) and Mn (e) atoms. The position of the atom is indicated by the orange circle. Scale bars are 2 nm. Grids of dI/dV spectra on these areas were fit with a Fano function combined with a gap as described in the text. For the fits, Γ_r was restricted to maximum 20 meV, q factor to $[-1; 1]$, and E_r $[-10; 10]$ meV. The extracted Fano parameters are shown for Fe (b)–(d) and Mn (f)–(h) adatoms (resonance width Γ_r , q factor, and resonance energy E_r). White pixels are used when the fits did not yield reasonable results. dI/dV spectra recorded with (b)–(d): $V_s = 40$ mV, $I_t = 300$ pA, $V_{\text{mod}} = 0.5$ mV, and (f)–(h): $V_s = 100$ mV, $I_t = 300$ pA, $V_{\text{mod}} = 1$ mV.

we also do not find significant correlations between them (see Appendix). This contrasts with the observation by Zhao *et al.* who found that the asymmetry was changing across a Fe2 vacancy [22]. In inspecting our topography, we note that most areas do not allow to identify isolated Fe2 vacancies, but a disordered modulation of the density of states, hinting at many different vacancies that may be located at different sample depths. This may preclude the observation of a direct correlation to specific topographic changes. However, we find a similar pattern, i.e., a direct correlation between the asymmetry parameter q and the position of the Fano resonance E_r (see Appendix for correlation map). This suggests that the local doping level determining the local electron-hole asymmetry is probed by the interfering tunneling paths.

Surprisingly, the addition of magnetic Fe or Mn atoms does not lead to any noticeable variation of the Kondo correlation strength Γ_r beyond the variations on the pristine background [see Figs. 3(b) and 3(f)]. Furthermore, the Fe atom seems to be transparent to any of the other Kondo parameters, neither affecting the position of the Kondo resonance, nor the asymmetry. This is contrasted by the influence of the Mn atom, which leads to a significant change in the asymmetry q of the resonance. The cotunneling paths contributing to interference are determined by the electronic structure of the adatom. Different orbital occupations, thus, modify the interference of the tunneling paths [38–43].

Next, we also investigate the spatial variation and response of the small gap to the presence of the Fe atom. Figure 4(a)

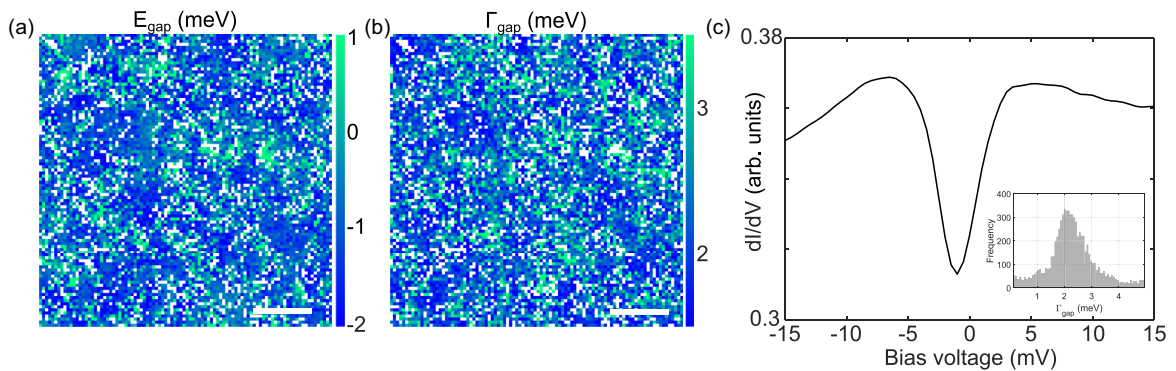


FIG. 4. Spatial variation of the hybridization gap. (a), (b) Extracted gap energy E_{gap} and gap size Γ_{gap} from the grid of spectra shown in Fig. 3(a). The spectra are fit by a function that combines a Fano line shape, a gap, and a linear background as described in the text. White pixels are used when the fits did not yield reasonable results. (c) Plot of the average of all remaining gaps after subtraction of the Fano line shape in the area of Fig. 3(a). The inset in Fig. 4(c) shows the histogram of the gap FWHM values. The Fano contribution of the fit is subtracted from the spectra. Scale bars are 2 nm.

shows the spatial variation of the gap energy and Fig. 4(b) the corresponding width that has been extracted from the combined fit with the gap and Fano functions. The gap persists everywhere on the surface albeit with spatial variations [see inset in Fig. 4(b)], reflecting the disorder already observed in topography. The average of the gap is shown in Fig. 4(c) and amounts to ≈ 2.3 meV. The histogram of the gap width [inset in Fig. 4(c)] shows a rather symmetric distribution around its mean value, suggesting the absence of multifractality [44]. The difference between our observation and the study of Mathimalar *et al.* [30] may be ascribed to a different defect density. Surprisingly, we do not find any impact of the Fe atom on the small hybridization gap.

The absence of any noticeable effect on the correlated state of the underlying substrate by magnetic adsorbates suggests that the intralayer correlations are robust against external impurities. The exchange coupling of a weakly adsorbed atom on the terminating Te layer is, thus, insufficient to perturb the stronger correlations within the layers, which themselves are prone to variations imposed by Fe vacancies.

III. CONCLUSIONS

We used scanning tunneling spectroscopy at 1.1 K to investigate the low-energy excitation spectra on the surface of Fe_3GeTe_2 . While we reproduced the previously observed Fano line shape on the energy scale of ≈ 10 meV, which has been interpreted as evidence of the Kondo effect, we additionally identified a small energy gap superimposed on this spectrum. Consistent with earlier predictions of heavy-fermion behavior, we attribute this gap to a hybridization gap arising from the long-range coherent interaction of Kondo impurities. These findings add to the debate regarding the presence of Kondo-lattice behavior, which had appeared incompatible with the simultaneous ferromagnetic state. Further evidence for a robust long-range correlated state in the substrate was provided by the spatial mapping of the Kondo-coupling parameters. The Kondo resonance as well as the hybridization gap persist all across the surface albeit with spatial variations that we ascribe to the inhomogeneous distribution of Fe vacancies in the substrate. While this suggests that magnetic properties can be tuned by the Fe content within the layer, magnetic adatoms on the surface do not significantly perturb the correlations within the layer, suggesting stronger intralayer interactions. Our findings provide additional evidence of the complex magnetic properties of Fe_3GeTe_2 and underscore the need to consider the contributions of distinct Fe sites to fully explain its magnetic behavior.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [45].

APPENDIX

The maps shown in Fig. 3 show that the variations in the Fano parameters occur in the same scale of the topographic height variations. To analyze this in detail, we calculated the correlation among topography, all Fano parameters, and the small gap parameters using the Pearson correlation coefficient r with

$$r = \frac{\sum_m \sum_n (A_{mn} - \bar{A})(B_{mn} - \bar{B})}{\sqrt{(\sum_m \sum_n (A_{mn} - \bar{A})^2)(\sum_m \sum_n (B_{mn} - \bar{B})^2)}} \quad (\text{A1})$$

where A_{mn} is the value of the first map in the positions (m, n) , B_{mn} is the value of the second map in the positions (m, n) , $\bar{A}$ and $\bar{B}$ are the mean values of map A and B. The results are summarized in the Table I. Only the energy position of the Fano resonance and the Fano asymmetry parameter q are correlated.

We also calculated the two-dimensional correlation of each map. The resulting maps are shown in Fig. 5.

TABLE I. Correlation values between the topography, Fano parameter, and gap parameter maps.

	Topo	q	Γ_r	E_r	Γ_{gap}	E_{gap}
Topo		0.3	-0.1	0.1	0.001	0.07
q			0.01	0.8	-0.01	0.05
Γ_r				0.05	0.01	-0.2
E_r					-0.01	-0.02
Γ_{gap}						-0.1
E_{gap}						

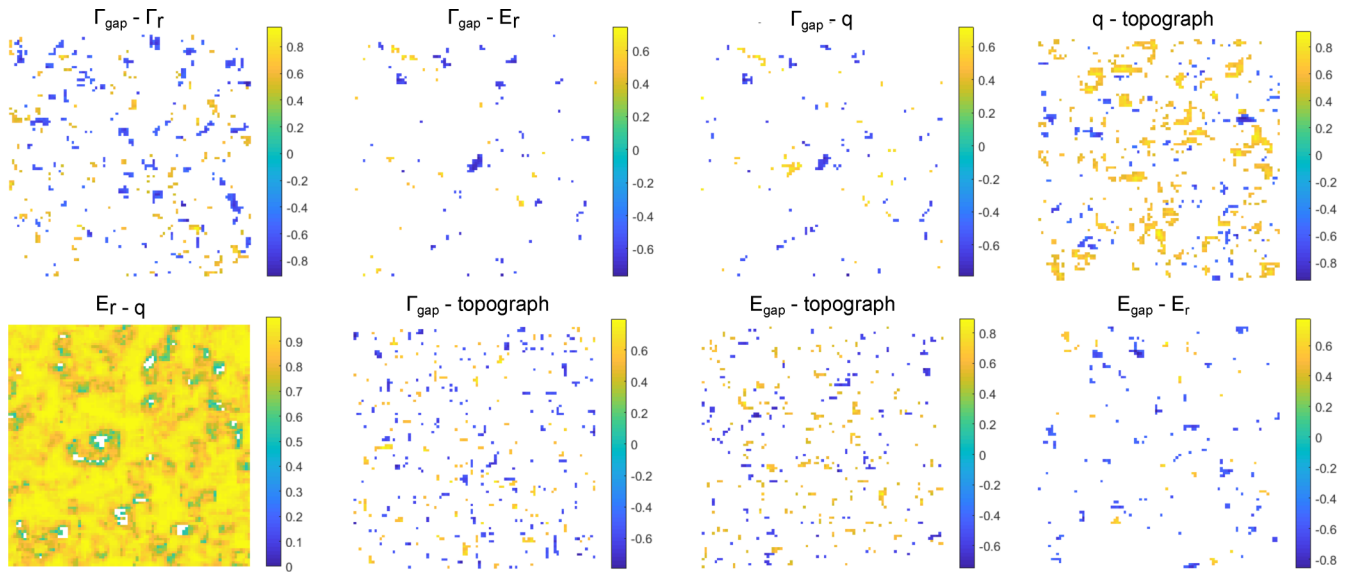


FIG. 5. Correlation maps for several gap and Fano parameters. For clarity, only values above $|0.5|$ correlation are shown. Empty pixels represent correlation values below $|0.5|$.

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