General, Strong Impurity-Strength Dependence of Quasiparticle Interference

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Quasiparticle interference patterns in momentum space are often assumed to be independent of the strength of the impurity potential when comparing with other quantities, such as the joint density of states. Here, using the $T$-matrix theory, we show that this assumption breaks down completely even in the simplest case of a single-site impurity on the square lattice with an $s$ orbital per site. Then, we predict from first principles a very rich, impurity-strength-dependent structure in the quasiparticle interference pattern of TaAs, an archetype Weyl semimetal. This study thus demonstrates that the consideration of impurity strength is essential for interpreting Fourier-transform scanning tunneling spectroscopy experiments in general.

Keywords: Quasiparticle interference, Scanning tunneling microscopy, Fourier-transform scanning tunneling spectroscopy, First-principles calculation, Wannier function, Impurity strength

Scanning tunneling microscopy plays a key role in nanoscience because it directly probes the surface topography and electronic density of states with a sub-nanometer resolution [1, 2]. In particular, the Fourier-transform scanning tunneling spectroscopy has been widely used to examine the surface electronic structures in momentum space [3]. The experimental results are interpreted as the result of quasiparticle interference (QPI) induced by impurities or defects on the surface. Theoretically, a QPI pattern is defined as the Fourier transform of the perturbation to the local density of states (LDOS) induced by the impurity [4]. The QPI patterns of various materials including Weyl semimetals [5–9], high-$T_c$ superconductors [10–14], and topological insulators [9, 15, 16] are being actively studied.

One often analyzes the observed QPI patterns by comparing them to the joint density of states (JDOS) [3]

$$ J(q;\omega) = \int dk \rho_0(k;\omega) \rho_0(k - q;\omega). \tag{1} $$

Here,

$$ \rho_0(k;\omega) = -\frac{1}{\pi} \text{Tr} \text{Im} G_0(k;\omega) \tag{2} $$

is the surface density of states at wavevector $k$ and energy $\omega$ in the absence of impurities with $G_0$ the surface Green function. Since there is no reference to the properties of the impurities in Eq. (1), the JDOS approximation neglects the impurity dependence of the QPI.

Another commonly used approximation of the QPI pattern is the spin scattering probability (SSP) [17]

$$ J_s(q;\omega) = \sum_{i=0}^3 \int dk \rho_i(k;\omega) \rho_i(k - q;\omega), \tag{3} $$

where

$$ \rho_i(k;\omega) = -\frac{1}{\pi} \text{Tr} \text{Im} \sigma_i G_0(k;\omega) \tag{4} $$

is the spin density along the $i$-th direction ($i = 1, 2, 3$) with $\sigma_i$ the Pauli matrix. Inoue et al. used surface-projected SSP to explain the measured QPI patterns in TaAs [5]. The SSP approximation goes beyond the JDOS approximation by forbidding the scatterings between oppositely-polarized pure spin states. Still, SSP is independent of impurity-specific properties.

The Born approximation is also frequently used to simulate QPI. Under the Born approximation, one takes into account only the first-order effect of the scattering potential on the Green function. Consequently, the QPI pattern computed within the Born approximation is independent of the strength of the scalar impurities up to an overall prefactor. In this paper, we use the $T$-matrix method [18] to investigate the effect of the strength of simple non-magnetic scalar impurities on the QPI pattern. Contrary to the common belief, the QPI pattern depends very sensitively on the impurity strength. Thus, the approximations that do not capture such impurity-

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**FIG. 1.** The structure of the systems examined in this paper. (a) Single-orbital square lattice. (b) As-terminated semi-infinite surface of TaAs.
strength dependence, such as JDOS, SSP, and the Born approximation, may fail to describe even the QPI arising from simple non-magnetic impurities. We first show that the QPI patterns of a simple square lattice [Fig. 1(a)] depend dramatically on the impurity strength. After fully understanding the physics of this simplest system, we look into TaAs [Fig. 1(b)], an archetype Weyl semimetal, and find that its QPI pattern also depends significantly on the impurity strength.

The Green function of a semi-infinite surface in the absence of impurities is defined as

$$G_0(\omega) = (\omega - i0^+ - \mathcal{H}_0)^{-1},$$

where $\mathcal{H}_0$ is the Hamiltonian of the pristine semi-infinite surface. The Green function of the pristine surface is block diagonal with respect to the in-plane wavevectors. The Green function for a wavevector $\mathbf{k}$ is

$$G_0(\mathbf{k}; \omega) = (\omega - i0^+ - \mathcal{H}_0 P_k \mathcal{H}_0 P_k^{-1})^{-1},$$

where $P_k$ is the projection operator onto the subspace with wavevector $\mathbf{k}$.

Now, let us introduce a scalar impurity with potential $V$. In this paper, we work using the tight-binding description of the Hamiltonian and consider the case where the impurity potential shifts the onsite energy of all orbitals of the topmost atom in the central in-plane unit cell. Concretely, the impurity potential matrix element for orbitals $m$ and $n$ in in-plane unit cells $\mathbf{R}$ and $\mathbf{R'}$ is

$$V_{m\mathbf{R},n\mathbf{R'}} = V(P_T)_{m,n} \delta_{\mathbf{R},0} \delta_{\mathbf{R'},0},$$

with $V$ the impurity strength. Here, $P_T$ is a projection operator to the orbitals in the topmost atom, so that $(P_T)_{m,n} = 1$ if $m = n$ and $m$ is an orbital that belongs to the topmost atom, and 0 otherwise.

Using the $T$-matrix formalism, the change in the Green function induced by the perturbation $V$ reads [3]

$$\Delta G(\omega) = G(\omega) - G_0(\omega) = G_0(\omega) T(\omega) G_0(\omega),$$

where the $T$ matrix is defined as

$$T(\omega) = V[I - G_0(\omega) V]^{-1},$$

with $I$ the identity operator. For the impurity potential defined in Eq. (7), the $T$ matrix becomes

$$T(\mathbf{R}, \mathbf{R'}; \omega) = T \delta_{\mathbf{R},0} \delta_{\mathbf{R'},0}$$

$$= V[P_T - P_T G_0(0, 0; \omega) P_T]^{-1} \delta_{\mathbf{R},0} \delta_{\mathbf{R'},0}$$

where the real-space Green function is defined as

$$G_0(\mathbf{R}, \mathbf{R'}; \omega) = \sum_{\mathbf{k}} e^{-i\mathbf{k} \cdot (\mathbf{R} - \mathbf{R'})} G_0(\mathbf{k}; \omega).$$

To simulate scanning tunneling spectroscopy, we project the Green function to the topmost atomic layer, assuming that the only the LDOS of the topmost atomic layer is measured. The change in the surface LDOS induced by the impurity reads

$$\Delta \rho(\mathbf{R}; \omega) = -\frac{1}{\pi} \text{Im} \sum_{i \in \text{topmost}} \langle w_{i\mathbf{R}} | \Delta G(\omega) | w_{i\mathbf{R}} \rangle,$$

where $| w_{i\mathbf{R}} \rangle$ is the localized orthogonal basis function, such as the Wannier function. The QPI pattern $\Delta \rho(\mathbf{q}; \omega)$ is the Fourier transform of $\Delta \rho(\mathbf{R}; \omega)$:

$$\Delta \rho(\mathbf{q}; \omega) = i \frac{1}{2\pi} \sum_{i \in \text{topmost}} \int d\mathbf{k} \langle w_{ik} | \Delta G - \Delta G^\dagger | w_{ik} \rangle.$$

If the system is invariant under a $C_2$ rotation with respect to $z$ [Fig. 1], Eq. (13) reduces to

$$\Delta \rho(\mathbf{q}; \omega) = -\frac{1}{\pi} \text{Im} \sum_{i \in \text{topmost}} \int d\mathbf{k} \langle w_{ik} | \Delta G | w_{ik} \rangle.$$

Using Eqs. (8, 10, 14) with the Green function of the pristine surface computed from the iterative method [19], one can efficiently compute the QPI pattern induced by a localized potential impurity.

Now, as the simplest example, let us consider the square lattice with one s-like orbital per site with nearest-neighbor hopping with hopping integral $t$. The energy dispersion reads

$$\epsilon_k = 2t [\cos(ak_x) + \cos(ak_y)],$$

FIG. 2. JDOS [(a)], absolute value of the QPI pattern [(b) and (c)], and absolute value of the QPI pattern computed within the Born approximation [(d)] for the single-orbital square lattice with $t = -1$ eV at $\omega = 3.5$ eV. The dashed curves are circles with radius $q = 2k_c$. 
where $a$ is the lattice parameter. In this single-orbital case, the $T$ matrix becomes a complex number and so is $G_0(k; \omega)$. The QPI pattern becomes

$$\Delta \rho(q; \omega) = -\frac{1}{\pi} \text{Im} [T \Pi(q; \omega)] \quad (16)$$

where

$$\Pi(q; \omega) = \int dk \ G_0(k; \omega) G_0(k - q; \omega). \quad (17)$$

Figure 2(a) and (b, c) show the computed JDOS and QPI patterns of the single-orbital square lattice with $t = -1$ eV, respectively. The quantities are calculated at $\omega = 3.5$ eV. At this energy, the constant-energy contour is approximately a circle with radius $k_c = 0.225 \pi / a$.

Comparing the JDOS and the QPI patterns, we find that the intensity outside the $q = 2k_c$ circle are clearly different: the JDOS is zero outside the $q = 2k_c$ circle, while the QPI patterns are non-zero. More importantly, we find a large difference between the QPI patterns for $V = 1$ eV and $V = 3$ eV.

Within the Born approximation, the $T$ matrix is replaced by the impurity potential $V$. Therefore, the computed QPI pattern becomes independent of the impurity strength $V$ except for a proportionality constant.

The QPI pattern computed within the Born approximation [Fig. 2(d)] is more similar to the exact QPI pattern for small $V$ [Fig. 2(b)] than that for large $V$ [Fig. 2(c)] since the Born approximation becomes more accurate for weaker perturbations. However, the QPI pattern of a stronger impurity [Fig. 2(c)] considerably deviate from the Born approximation.

To understand the impurity-strength dependence of the QPI patterns shown in Fig. 2, we examine the $T$ matrix. Figure 3(a) shows the real and imaginary parts of $T$ as a function of $V$. Also, we plot $T$ at $V = \pm \infty$, which can be used to simulate a vacancy at the impurity site. At small $|V|$, the real part of $T$ can be approximated as $V$ and its imaginary part is negligible, indicating the adequacy of the Born approximation. However, at large $|V|$, both $\text{Re} T$ and $\text{Im} T$ are sizable.

The QPI signal is proportional to the imaginary part of $T \Pi$ [Eq. (17)]. Thus, only the imaginary part of $\Pi$ contribute to the QPI pattern at small $|V|$, while both $\text{Im} \Pi$ and $\text{Re} \Pi$ contributes at larger $|V|$.

Using the obtained $T$ matrix, we now aim to understand the $V$ dependence of the QPI patterns [Fig. 2(b) and (c)]. In Fig. 3(b), we show the absolute value of the QPI patterns along the line $q_y = 0$. This seemingly complicated dependence of the QPI pattern on $V$ can be simply understood by comparing the signed QPI patterns [Fig. 3(c)] and the $\Pi$ function [Eq. (17) and Figs. 3(d,e)] (see also Fig. S1 [20]). The complementary feature of $\text{Re} \Pi$ and $\text{Im} \Pi$ is the key to understanding the impurity-strength dependence of the QPI pattern. Using Eq. (16), one can write the QPI signal as

$$\Delta \rho(q; \omega) = -\frac{1}{\pi} [\text{Im} T \ \text{Re} \Pi(q; \omega) + \text{Re} T \ \text{Im} \Pi(q; \omega)].$$

(A18)

Apparently, the QPI signal is a linear combination of $\text{Re} \Pi$ and $\text{Im} \Pi$ with coefficients $-\frac{1}{\pi} \text{Im} T$ and $-\frac{1}{\pi} \text{Re} T$, respectively. For $V = 1$ eV, since $|\text{Im} T| < |\text{Re} T|$ [Fig. 3(a)], $\text{Im} \Pi$ dominates the qualitative features of the QPI pattern. However, for $V = 3$ eV, we find $|\text{Im} T| \gg |\text{Re} T|$ [Fig. 3(a)], and thus the QPI pattern is almost exclusively determined by $\text{Re} \Pi$. This analysis clearly demonstrates why the signed QPI patterns for $V = 1$ eV and $V = 3$ eV [Fig. 3(c)] resemble $-\text{Im} \Pi$ [Fig. 3(e)] and $-\text{Re} \Pi$ [Fig. 3(d)], respectively. In brief, the position of peaks are determined by $\Pi$ or the
Green function $G_0(k;\omega)$ [Eq.(17)], while their intensity and shape are determined by $T$.

In passing, we note that Ref. [21] investigated the impurity-strength dependence of the QPI patterns of the parent compounds of iron-pnictide superconductors, magnetic materials with a spin density wave order. The subjects and findings of our work and Ref. [21] are totally different as in that work, the peaks in the energy position of the spin-density wave are of crucial importance. We also note that the findings in our work are relevant to general, even non-magnetic systems.

In order to see the effect of real-space masking performed as a post-processing of the experimental data [15], we applied the real-space masking to our calculated QPI patterns. We find that the real-space masking still preserves much of the impurity-strength dependence of the QPI patterns (see Fig. S2 [20]).

We now move on to the As-terminated surface of TaAs. We used the Quantum ESPRESSO package [22, 23] for density-functional theory calculations and the Wannier90 package [24, 25] to construct the ab initio Wannier-function-based tight-binding models. (See Supplementary Information for the computational details [20].) When computing the JDOS [Eq. (1)], the SSP [Eq. (3)], and the QPI pattern [Eq. (13)], the sum over atomic orbitals was limited to the orbitals belonging to the topmost As atoms.

The SSP and the QPI patterns of TaAs are shown in Fig. 4. The JDOS (see Fig. S3 and S4 [20]) and SSP have only minor differences [5]. However, as in the case of the square lattice (Fig. 2), the QPI pattern is widely different from the JDOS and SSP. Moreover, the calculated QPI patterns are strongly dependent on the impurity strength. We note that a direct comparison between our results and the theoretical results in Ref. [5] cannot be made because the Umklapp scattering processes were neglected therein. We also note that the QPI patterns of TaAs calculated in our study cannot be directly compared with the experimental results in Ref. [5]. The purpose of our study is to show that the QPI patterns depend strongly on the impurity strength even for the simplest impurities and not to find the exact impurity potential for a specific experimental study [5] in which the kind of impurities is not investigated.

To understand this strong impurity-strength dependence of the QPI patterns, we investigated the corresponding signed QPI signals. Figure 5 shows that the positions of the peaks of $\Delta \rho$ are rather insensitive to $V$ and are determined by the electronic structures of TaAs, as similar peaks also occur in the SSP (or JDOS; see Fig. S4 [20]). However, the signed QPI patterns vary strongly with $V$. These behaviors are very similar to the case of the square lattice [Fig. 3(c)]. Since the impurity potential acts on multiple orbitals of the topmost As atom, the $T$ matrix is now a matrix, not a complex number. Hence, a simple analysis like Eq. (18) is not possible. However, as in the square lattice case, the position of the peaks are mainly determined by the pristine Green functions. Also, the impurity-strength dependence of the $T$ matrix is the origin of the complex impurity-strength dependence of the QPI patterns shown clearly in Fig. 4.

In conclusion, we have shown that the QPI pattern strongly depends on the strength of the impurity potential. This finding holds even in the simplest case of the square lattice with non-magnetic, scalar onsite impurities. The impurity-strength dependence is also present in the QPI patterns of TaAs. We were able to understand the complex impurity-strength dependence of the QPI patterns of both systems from a unified framework: the pristine surface Green functions determines the position of the peaks, while the intensity and shape of the peaks are significantly affected by the $T$ matrix. Our findings that the QPI patterns can be completely different for different types of impurities are general, with their profound applicability ranging from the simplest toy model to complicated topological materials. Therefore, our findings should generally be used in analyzing the results of Fourier-transform scanning tunneling spectroscopy experiments.

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FIG. 4. SSP [(a)], and the absolute value of the QPI patterns [(b)-(e)] for the As-terminated TaAs surface for $\omega$ at the Fermi level.

FIG. 5. Signed QPI patterns for As-terminated TaAs surface along $q_y = 0$ for $\omega$ at the Fermi level. We note that $\Delta \rho(q; \omega)$ is real valued [Eq. (14)].

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General, Strong Impurity-Strength Dependence of Quasiparticle Interference: Supplementary Information

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FIG. S1. Real [(a)] and imaginary [(b)] parts of \( \Pi \) [Eq. (18) of the main manuscript] for the square lattice with \( t = -1 \) eV at \( \omega = 3.5 \) eV. The dashed curves are circles with radius \( q = 2k_c \).

GREEN FUNCTION CALCULATION

We applied the iterative scheme to obtain the surface Green function for the pristine semi-infinite surface [1]. For the calculation of the QPI patterns, in the case of the single-orbital square lattice, we used a 200 \( \times \) 200 k-point grid and a 0.05 eV broadening. In the case of TaAs surface, we used a 400 \( \times \) 400 k-point grid and a 0.01 eV broadening.

REAL-SPACE MASKING

Figure S2 shows the effects of real-space masking on the QPI patterns. Figure S2 (b), (c), (e), and (f) are obtained by Fourier transforming the masked LDOS variation \( \Delta \rho_{\text{masked}}(\mathbf{R}; \omega) \) instead of the unmasked one \( \Delta \rho(\mathbf{R}; \omega) \). For the Gaussian masking, the masked LDOS variation is

\[
\Delta \rho_{\text{masked}}(\mathbf{R}; \omega) = \Delta \rho(\mathbf{R}; \omega) \times [1 - \exp(-R^2/\eta^2)].
\]

For the cutoff masking, we set

\[
\Delta \rho_{\text{masked}}(\mathbf{R}; \omega) = \Delta \rho(\mathbf{R}; \omega) \times \Theta(R - \eta)
\]

with \( \Theta(x) \) the Heaviside step function.

DETAILS OF FIRST-PRINCIPLES CALCULATIONS

We used the Quantum ESPRESSO package [2, 3] for the density functional theory computation with a plane-wave basis set. The kinetic-energy cutoff of 70 Ry was used. The parametrization of Perdew, Burke, and Ernzerhof (PBE) was used for the generalized-gradient approximation of the exchange-correlation functional [4]. We included spin-orbit coupling by using fully relativistic pseudopotentials. Fully relativistic pseudopotentials for Ta and As were taken from the SG15 library [5–7]. Magnetism was not considered. We applied the Methfessel-Paxton smearing [8] of 0.01 Ry. The k-point grid for the bulk calculation was set to 12 \( \times \) 12 \( \times \) 8. For the slab calculation, we used a slab with 40 atomic layers and used a 12 \( \times \) 12 \( \times \) 1 k-point grid. We used the experimental lattice parameters of TaAs [9]. We relaxed the atomic coordinates of the bulk until the forces on the atoms were below \( 2 \times 10^{-3} \) eV/Å. We did not relax the structure of the slab.

To generate the Wannier-function-based tight-binding Hamiltonian, we used the Wannier90 package [10, 11]. We used the projection-only Wannier functions, which are generated without any iterative localization procedures. Consequently, the bulk and slab tight-binding models could be stitched to a semi-infinite surface model without any corrections [12]. For Wannierization, the Brillouin zone was sampled with uniform 7 \( \times \) 7 \( \times \) 6 and 7 \( \times \) 7 \( \times \) 1 grids for the bulk and slab, respectively. We used the Ta-centered \( d \) orbitals and As-centered \( p \) orbitals as the initial guesses for the Wannier functions. The spinor parts of the initial guesses were aligned along the \( z \) axis.

The inner (frozen) windows were set to [-2, 1] eV around the Fermi level for the bulk and slab. The outer (disentangled) windows were set to [-10, 9] eV and [-9.5, 9.5] eV around the Fermi level for the bulk and slab, respectively. The ab initio tight-binding Hamiltonian is symmetrized by zeroing out a hopping integral if any of its symmetry-equivalent hopping integrals is not included in the tight-binding Hamiltonian due to the finite Brillouin zone sampling.

In Fig. S3, we plot the surface LDOS of the TaAs surface. There, the surface Fermi arcs are observed. The computed surface LDOS agrees with previous surface-
FIG. S2. Effects of the real-space masking on the QPI patterns of the square lattice with $t = -1$ eV at $\omega = 3.5$ eV. (a) and (d): Unmasked QPI patterns. (b) and (e): Gaussian masked QPI patterns, (c) and (f): Cutoff masked QPI patterns. The dashed curves are circles with radius $q = 2k_c$.

FIG. S3. Fourier transform of the LDOS at the orbitals localized at the topmost As atom for the As-terminated TaAs surface at the Fermi level.

FIG. S4. Surface-projected JDOS for the orbitals localized at the topmost As atom for the As-terminated TaAs surface for $\omega$ at the Fermi level.

specific calculations [13–15].

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FIG. S5. Signed values of the QPI signal for the As-terminated TaAs surface along the $q_y = 0$ [(a)], $q_x = 0$ [(b)], and $q_x = q_y$ [(c)] lines.

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