Quantifying many-body effects by high-resolution Fourier transform scanning tunneling spectroscopy

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Many-body phenomena are ubiquitous in solids, as electrons interact with one another and the many excitations arising from lattice, magnetic, and electronic degrees of freedom. These interactions can subtly influence the electronic properties of materials ranging from metals,1 exotic materials such as graphene,2,3 and topological insulators,4 or they can induce new phases of matter, as in conventional5 and unconventional superconductors,6–9 heavy fermion systems,10 and other systems of correlated electrons. As no single theoretical approach describes all such phenomena, the development of versatile methods for measuring many-body effects is key for understanding these systems. To date, angle-resolved photoemission spectroscopy (ARPES) has been the method of choice for accessing this physics by directly imaging momentum resolved electronic structure.2–4,6,7,9 Scanning tunneling microscopy/spectroscopy (STM/S), renown for its real-space atomic resolution capability, can also access the electronic structure in momentum space using Fourier transform scanning tunneling spectroscopy (FT-STS).11–14 Here, we report a high-resolution FT-STS measurement of the Ag(111) surface state, revealing fine structure in the otherwise parabolic electronic dispersion. This deviation is induced by interactions with lattice vibrations and has not been previously resolved by any technique. This study advances STM/STS as a method for quantitatively probing many-body interactions. Combined with the spatial sensitivity of STM/STS, this technique opens a new avenue for studying such interactions at the nano-scale.
Non-interacting electrons in a crystal occupy quantum states with an infinite lifetime and band dispersion $\epsilon(k)$ set by the lattice potential. Interactions with the other electrons and elementary excitations of the system scatter the electrons, resulting in an altered dispersion relation $E(k)$ and a finite lifetime. These many-body effects are encoded in the complex self-energy $\Sigma(k, E) = \Sigma'(k, E) + i\Sigma''(k, E)$. The imaginary part $\Sigma''(k, E)$ determines the lifetime of the state and is related to the scattering rate. The real part $\Sigma'(k, E)$ shifts the electronic dispersion $E(k) = \epsilon(k) + \Sigma'(k, E)$. The tools available for studying energy and momentum resolved self-energy are limited. For example, bulk transport and optical spectroscopies provide some access to $k$-integrated self-energies while ARPES accesses $k$-resolved information for only the occupied states. It is therefore important to develop a more extensive suite of versatile techniques, especially in the context of complex systems that remain poorly understood from a theoretical perspective.

STM/STS accesses momentum space electronic structure by imaging real-space maps of the modulations in differential conductance $(dI/dV)$, which is proportional to the local density of states (LDOS) of the sample. These modulations arise from the interference of electrons scattered elastically by defects, and contain information about the initial and final momenta that are accessible by a Fourier transform of the real space map. As the electrons are dressed by interactions, the momentum space scattering intensity map is often referred to as the quasiparticle interference (QPI) map. The dominant intensities in a QPI map occur at scattering wave vectors linking constant energy segments of the band dispersion. By tracking the energy dependence of these peaks, the electronic dispersion $E(k)$ can be obtained. This technique has been used to map coarse dispersions in many materials and to examine scattering selection rules. While the influence of many-body effects in FT-STS has been postulated since early reports, the direct influence on dispersion and a quantitative account of self-energy effects were missing. Here, we extend this technique to the examination of fine structure in the $E(k)$ due to many-body interactions.

The two-dimensional Shockley surface state of the noble metal silver Ag(111) was selected as an ideal system for demonstrating the quantitative capabilities of FT-STS. The Ag(111) surface state is well characterized, and while it exhibits distinct many-body effects, it lacks the complicated
interplay of interactions that appear in many complex modern materials. The dispersion of the surface state is free-electron-like over a wide energy range given by \( \epsilon(k) = \hbar^2 k^2 / 2m^* - \mu \), where \( m^* \) is the effective mass and \( \mu \) is the chemical potential. This parabolic dispersion is modified by the electron-electron (e-e) and electron-phonon (e-ph) interactions. As both are accurately described by conventional theory, a straightforward comparison with theory can be made, requiring few parameters. The e-e interaction decreases the electron lifetime for energies away from the Fermi level. The e-ph interaction introduces an additional scattering channel for energies above the typical phonon energy scale (the Debye frequency \( h\Omega_D \)), decreasing the lifetime and modifying the bare dispersion near the Fermi energy \( E_F \). The latter is a subtle effect in the case of Ag(111), that had not yet been observed due to the high resolution required in both energy and momentum.

STM/STS measurement of the Ag(111) surface yields real-space conductance maps (see Fig. 1a for the map at \( E = E_F \)) with circular LDOS modulations arising from scattering from point-like CO adsorbates, and vertical modulations produced by step-edge reflections. The small terraces on the surface produce subtle confinement effects not representative of the pristine surface state. In order to access intrinsic surface properties, we removed their contribution by setting \( dI/dV \) in this region to the average value over the entire image. The ability to isolate regions of interest in this way is unique to STM/STS, as probes such as ARPES would average over these domains. Fig. 1b shows a typical \( dI/dV \) spectra, averaged over a defect free region. The momentum space QPI intensity map \( S(q, E_F) \) (Fig. 1c) exhibits a ring of radius \( q(E_F) = 2k(E_F) \), as expected for a free-electron-like dispersion where back scattering is dominant. A line profile \( S(|q|, E_F) \) of the QPI map is shown in Fig. 1d, where we have performed an angular average of \( S(q, E_F) \) in the regions above and below the dashed lines. This restriction isolates the contributions of the point-like CO scatterers. The momentum space resolution \( \Delta q \sim 0.0026 \, \text{Å}^{-1} \) is set by the dimension of the map (239 × 239 nm\(^2\)) while the energy resolution \( \Delta E = 4k_B T = 1.5 \, \text{meV} \) is limited by thermal broadening of the tip and the sample.

We now examine the detailed electronic structure of the Ag(111) surface state by consider-
ing the full energy dependence of the angle-averaged profile $S(|\mathbf{q}|, E)$, as shown in Fig. 2a. A parabolic band dispersion is evident over a wide energy range while the QPI signal intensity exhibits a monotonic decrease from the onset of the surface state to higher energy. From the data we extract $\mu = 65 \pm 1$ meV and $m/m_e = 0.41 \pm 0.02$, consistent with previous measurements.\cite{18,20,23,24}

These values define the bare dispersion in the absence of e-ph coupling. Deviations from this dispersion, as well as an enhanced QPI signal intensity, are evident in the vicinity of $E_F$, shown more clearly in the inset of Fig. 2a. A similar increase in QPI signal intensity was observed near $E_F$ in one of the first FT-STS reports on the Be(0001) surface state\cite{11} and the e-ph interaction was later proposed as a possible origin.\cite{29}

To identify the source of these deviations and to assess the QPI signal we modeled the system using the T-matrix formalism, considering scattering from a single CO impurity in the unitary limit (see methods). The e-e interaction was handled within Fermi liquid theory while the e-ph interaction was treated within standard Migdal theory\cite{1} with the phonons described by the Debye model (Debye frequency $\hbar \Omega_D = 14$ meV, dimensionless e-ph coupling strength $\lambda = 0.13$). Within these approximations the self-energy $\Sigma(E)$ is a function of energy only. The resulting simulated QPI intensity is shown in Fig 2b. The model closely reproduces both the coarse and fine details of the data. The overall monotonic decrease in QPI intensity is linked to the group velocity of the bare electronic dispersion and is not related to many-body effects. However, the increase in the intensity near $E_F$ and the deviations from the parabolic band dispersion arise from the e-ph interaction.

We now perform a quantitative analysis of the self-energy. For reference, Fig. 3a shows the e-ph self-energy for the same values of $\hbar \Omega_D$ and $\lambda$ used in Fig. 2b. To extract $\Sigma(E)$, a Lorentzian was fit to the data in the vicinity of the peak to obtain both the QPI peak position and height. (See methods. An example fit is shown as the dashed line in Fig. 1d.) A plot of the QPI peak height reflects the behavior of $\Sigma''(E)$ as shown in Fig. 3b, where we compare the data with the model. Here we have normalized both sets of data (as described in the figure caption) in order to eliminate the role of the tunneling matrix element in setting the scale of the experimental data. There is
good agreement between the model and experiment apart from a slight deviation $\sim 20$ meV, which we attribute to a set point effect below $q_F = 2k_F^{23}$. The decrease in $-\Sigma''(E)$ within $\hbar \Omega_D = 14$ meV of $E_F$ produces the non-monotonic variation in peak height superimposed over the group velocity dependence. This is due to the closing of the phonon scattering channel at energies below the characteristic phonon frequency, resulting in longer-lived quasiparticles near $E_F$. We note that the value $\hbar \Omega_D$ required to reproduce the data is close to the value for the top of the bulk acoustic branches$^{23}$. The real part $\Sigma'(E)$ can be estimated from the data by taking the difference between the measured peak position and the parabolic dispersion. The result is shown in Fig. 3c, where peaks in $\Sigma'(E)$ occur in the data at the same energy scale reflected in Fig. 3b. The dimensionless strength of the e-ph coupling $\lambda$ can be estimated from $d\Sigma'/dE\big|_{E=E_F}$. We obtain $\lambda = 0.13 \pm 0.02$, consistent with previous estimates$^{21,23}$.

Our FT-STS results provide a stunning visualization of the subtle modifications in dispersion and scattering intensity arising from many-body interactions in a simple system. This method provides high resolution in both momentum and energy that is competitive with state-of-the-art ARPES. Moreover, FT-STS accesses both occupied and unoccupied states opening up the possibility of examining particle-hole asymmetric systems. These aspects give access to many-body features not previously observed in such a direct way. With enhanced stability and lower temperatures, further advancements in the application of FT-STS to quantify many-body interactions in more complex systems can undoubtedly be expected. However, perhaps the most compelling advantage of FT-STS is the prospect of exploiting STM’s unique spatial sensitivity to explore variations in the many-body interactions in nanoscale regions and intrinsically inhomogeneous materials.

Methods

**Experiment:** Measurements were performed in a Createc STM in ultra high vacuum at a temperature of 4.2 K with a tungsten tip formed by direct contact with the Ag crystal. The Ag(111) surface
was cleaned by three cycles of Ar sputtering each followed by thermal annealing to 500 °C. The $I(V)$ map measured over 80 hours consists of $380 \times 380$ spectra taken on a $239 \times 239$ nm$^2$ area. Each $I(V)$ spectrum consists of 512 data points and was Gaussian smoothed maintaining thermally limited energy resolution of $\Delta E = 1.5$ meV. $dI/dV$ spectra were acquired by numerical differentiation of the $I$-$V$ sweep. Atomic resolution scans on the same area lead to a spatial resolution of $(0.58 \pm 0.01)$ nm. The peak position $S_O$ of each line profile $S(q, E)$ was determined by fitting a Lorentzian within a range of $\pm 0.01$ Å$^{-1}$ peak position expected from $\epsilon(k)$. The renormalization of the bare dispersion has been observed in four different data sets.

**Theory:** Calculations were performed using the $T$-matrix formalism for scattering from a single impurity$^{30}$ The QPI intensity is given by the impurity-induced LDOS modulations

$$
\delta \rho(q, \omega) = -\frac{i}{2\pi} \sum_k \text{Im}G(k, \omega)T(\omega)G(k + q, \omega).
$$

Here $T = -V_0 \sin(\delta) \exp(i\delta)$ is the $T$-matrix, where $\delta = \pi/2$ is the phase shift (unitary limit), $V_0$ is the scattering potential. The Green’s function is given by $G(k, \omega) = [\omega - \xi(k) - \Sigma(\omega)]^{-1}$ where $\xi = -\hbar^2 k^2/2m^* - \mu$ ($m^* = 0.41m_e, \mu = 65$ meV) is the dispersion of the surface state and $\Sigma(\omega) = -i\eta - i\gamma\omega^2/2 + \Sigma_{e-ph}(\omega)$ is the self-energy. Here $\eta = 2.5$ meV is the lifetime broadening due to scattering from the terraces, $\gamma = 62.7$ meV is the e-e interactions$^{24}$ and $\Sigma_{e-ph}(\omega)$ is obtained from a Debye model with $\hbar\Omega_D = 14$ meV and coupling strength $\lambda = 0.13^{24}$.
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Figure 1: A summary of FT-STS of the surface state of Ag(111). (a) Conductance map (dI/dV) of a 239 × 239 nm² area at $E = eV = 0$ meV (tip height set at $V = 100$ meV, $I = 200$ pA). LDOS modulation due to scattering at step edges and CO adsorbates are visible. The areas around the step edges were removed as discussed in the text. Furthermore, a tip change induced stripe was corrected by a line-by-line subtraction of the average line value that excludes CO impurities. (b) Average $dI/dV$ spectrum from a defect free area with a total size of 100 nm². The particle-hole symmetric steps at $E_F$ likely originate from an inelastic co-tunneling pathway via phonon modes polarized perpendicular to the surface. (c) Absolute value of the Fourier transform (power spectrum) of the $dI/dV$ map ($E = 0$, panel a) showing a ring with radius $q = 2k_F$, where $q$ is the scattering vector. The increased intensity along the $q_x$ direction originates from the step edge contributions. (d) The QPI line profile $S(q, E = 0)$. This is obtained by integrating (c) within the range above and below the dashed lines in order to isolate contributions from the CO adsorbates. The scattering peak is slightly asymmetric with an enhanced intensity at low $q$, which is more pronounced at higher energies. Scattering peak positions and heights were obtained by fitting the line profiles as shown in (a) within a range of $±0.01 \text{Å}^{-1}$ around the peak position (dashed line).
Figure 2: Measured and calculated dispersion of the QPI intensity. (a) The measured dispersion of $S(q, E)$, obtained by plotting the line profiles as shown in 1d for all bias voltages. Overall the dispersion is parabolic with $\mu = 65 \pm 1\,$meV and $m^*/m_e = 0.41 \pm 0.02$, obtained by fitting the peak position excluding the energy range $[-20, 20]\,$meV. The intensity of the scattering peak generally decreases with increasing energy but has a non-monotonic increase near $E_F$. We observe an additional scattering intensity below the onset of the surface state ($E < -70\,$meV). The inset reveals a subtle renormalization of the dispersion within $E_F \pm 14\,$meV. (b) Theoretical calculation of the QPI intensity with $\mu = 65\,$meV, $m^*/m_e = 0.41$. The model includes electron-electron (Fermi-liquid theory) and electron-phonon (Debye model, $\hbar\Omega_D=14\,$meV, $\lambda = 0.13$) interactions and assumes the CO adsorbates scatter in the unitary limit.
Figure 3: **Quantitative extraction of the real and imaginary part of the self-energy.** (a) Calculated real ($\Sigma'$, solid line) and imaginary ($\Sigma''$, dashed line) parts of the self-energy for the parameters used in Fig. 2b. (b) Scattering peak height $S_0(q, E)$ as a function of energy determined experimentally (dots) and theoretically (solid line). The theory curve has been normalized to the value at $E = 0$ while the experimental data had been normalized to the average value over the window $[-5, 5]$ meV. The overall decrease of the scattering peak height with increasing energy is related to the energy dependence of the group velocity. The increased intensity at $E_F \pm 14$ meV is caused by a dip in $-\Sigma''$ due to the e-ph interaction. This relationship opens up a new way to experimentally obtain $\Sigma''$. (c) Real part of the self energy determined from the difference between the scattering peak position $E(k)$ and a parabolic fit $\epsilon(q)$. The solid line corresponds to the theoretically determined $\Sigma'$ as presented in (a). Calculated and measured self-energy real part agree well, demonstrating that FT-STS is an alternative tool to obtain $\Sigma'(\omega)$. The dashed lines at $\pm 14$ meV in (a) to (c) indicate the position of the Debye energy $\hbar \Omega_D$. 
1. A. Migdal, “Interaction between electrons and lattice vibrations in a normal metal”. Sov. Phys. JETP 7, 996 (1958).

2. A. Bostwick, T. Ohta, T. Seyller, K. Horn, and E. Rotenberg, “Quasiparticle dynamics in graphene”. Nature Physics 3, 36 (2006).

3. A. Bostwick, F. Speck, T. Seyller, K. Horn, M. Polini, R. Asgari, A. H. MacDonald, and E. Rotenberg, “Observation of Plasmarons in Quasi-Freestanding Doped Graphene”. Science 328, 999 (2010).

4. Xuetao Zhu, L. Santos, C. Howard, R. Sankar, F. C. Chou, C. Chamon, and M. El-Batanouny, “Electron-Phonon Coupling on the Surface of the Topological Insulator Bi$_2$Se$_3$ Determined from Surface-Phonon Dispersion Measurements”. Phys. Rev. Lett. 108, 185501 (2012).

5. W. L. McMillan and J. M. Rowell, “Lead Phonon Spectrum Calculated from Superconducting Density of States”. Phys. Rev. Lett. 14, 108 (1965).

6. A. Lanzara, P. Bogdanov, X. Zhou, S. Kellar, D. Feng, E. Lu, T. Yoshida, H. Eisaki, A. Fujimori, K. Kishio, et al., “Evidence for ubiquitous strong electron-phonon coupling in high-temperature superconductors”. Nature 412, 510 (2001).

7. T. Dahm, V. Hinkov, S. Borisenko, A. Kordyuk, V. Zabolotnyy, J. Fink, B. Büchner, D. Scalapino, W. Hanke, and B. Keimer, “Strength of the spin-fluctuation-mediated pairing interaction in a high-temperature superconductor”. Nature Physics 5, 217 (2009).

8. K. Byczuk, M. Kollar, K. Held, Y.-F. Yang, I. Nekrasov, T. Pruschke, and D. Vollhardt, “Kinks in the dispersion of strongly correlated electrons”. Nature Physics 3, 168 (2007).

9. J. Graf, G.-H. Gweon, K. McElroy, S. Zhou, C. Jozwiak, E. Rotenberg, A. Bill, T. Sasagawa, H. Eisaki, S. Uchida, et al., “Universal High Energy Anomaly in the Angle-Resolved Photoemission Spectra of High Temperature Superconductors: Possible Evidence of Spinon and Holon Branches” Phys. Rev. Lett. 98, 067004 (2007).

10. Z. Fisk, H. Ott, T. Rice, and J. Smith, “Heavy electron metals”. Nature 320, 124 (1986).
11. P. Sprunger, L. Petersen, E. Plummer, E. Lgsgaard, and F. Besenbacher, “Giant Friedel Oscilla-
ations on the Beryllium(0001) Surface”. Science 275, 1764 (1997).

12. L. Petersen, P. T. Sprunger, P. Hofmann, E. Lgsgaard, B. G. Briner, M. Doering, H.-P. Rust, A.
M. Bradshaw, F. Besenbacher, and E. W. Plummer, “Direct imaging of the two-dimensional
Fermi contour: Fourier-transform STM”. Phys. Rev. B 57, R6858 (1998).

13. K. McElroy, R. Simmonds, J. Hoffman, D.-H. Lee, J. Orenstein, H. Eisaki, S. Uchida, and
J. Davis, “Relating atomic-scale electronic phenomena to wave-like quasiparticle states in
superconducting Bi$_2$Sr$_2$CaCu$_2$O$_{8+δ}$”. Nature 422, 592 (2003).

14. J. E. Hoffman, K. McElroy, D.-H. Lee, K. M. Lang, H. Eisaki, S. Uchida, and J. C. Davis,
“Imaging Quasiparticle Interference in Bi$_2$Sr$_2$CaCu$_2$O$_{8+δ}$”. Science 297, 1148 (2002).

15. J. P. Carbotte, T. Timusk, and J. Hwang, “Bosons in high-temperature superconductors: an
experimental survey”. Rep. Prog. Phys. 74, 066501 (2011).

16. G. M. Rutter, J. N. Crain, N. P. Guisinger, T. Li, P. N. First, and J. A. Stroscio, “Scattering and
interface in Epitaxial Graphene”. Science 317, 219 (2007).

17. P. Roushan, J. Seo, C. V. Parker, Y. Hor, D. Hsieh, D. Qian, A. Richardella, M. Z. Hasan, R.
Cava, and A. Yazdani, “Topological surface states protected from backscattering by chiral spin
texture”. Nature 460, 1106 (2009).

18. J. Li, W.-D. Schneider, and R. Berndt, “Local density of states from spectroscopic scanning-
tunneling-microscope images: Ag(111)”. Phys. Rev. B 56, 7656 (1997).

19. J. Li, W.-D. Schneider, R. Berndt, O. R. Bryant, and S. Crampin, “Surface-State Lifetime
Measured by Scanning Tunneling Spectroscopy” Phys. Rev. Lett. 81, 4464 (1998).

20. R. Paniago, R. Matzdorf, G. Meister, and A. Goldmann, “Temperature dependence of
Shockley-type surface energy bands on Cu(111), Ag(111) and Au(111)”. Surface Science 336,
113 (1995).
21. A. Eiguren, B. Hellsing, F. Reinert, G. Nicolay, E. Chulkov, V. Silkin, S. Hüfner, and P. Echenique, “Role of Bulk and Surface Phonons in the Decay of Metal Surface States”. Phys. Rev. Lett. 88, 066805 (2002).

22. L. Bürgi, L. Petersen, H. Brune, and K. Kern, “Noble metal surface states: deviations from parabolic dispersion”. Surface Science 447, L157 (2000).

23. L. Bürgi, H. Brune, O. Jeandupeux, and K. Kern, “Quantum coherence and lifetimes of surface-state electrons”. Journal of Electron Spectroscopy and Related Phenomena 109, 33 (2000).

24. L. Bügri, “Scanning tunneling microscopy as local probe of electron density, dynamics and transport at metal surfaces”. Ph.D. Thesis (1999).

25. R. Paniago, R. Matzdorf, G. Meister, and A. Goldmann, “High-resolution photoemission study of the surface states near $\bar{\Gamma}$ on Cu(111) and Ag(111)”. Surface Science 331, 1233 (1995).

26. J. Kliewer, R. Berndt, E. Chulkov, V. Silkin, P. Echenique, and S. Crampin, “Dimensionality Effects in the Lifetime of Surface States”. Science 288, 1399 (2000).

27. Y. Pennec, W. Auwärter, A. Schiffrin, A. Weber-Bargioni, A. Riemann, and J. Barth, “Supramolecular gratings for tuneable confinement of electrons on metal surfaces”. Nature Nanotechnology 2, 99 (2007).

28. L. Chen, L. L. Kesmodel, and J.-S. Kim, “EELS studies of surface phonons on Ag(111)”. Surface Science 350, 215 (1996).

29. M. Hengsberger, D. Purdie, P. Segovia, M. Garnier, and Y. Baer, “Photoemission Study of a Strongly Coupled Electron-Phonon System”. Phys. Rev. Lett. 83, 592 (1999).

30. L. Capriotti, D.J. Scalapino, and R.D. Sedgewick, “Wave-vector power spectrum of the local tunnelling density of states: ripples in a d-wave sea”. Phys. Rev. B 68, 014508 (2003).

**Competing interests statement**  The authors declare that they have no competing financial interests.