Origin of the crossover from polarons to Fermi liquids in transition metal oxides

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Transition metal oxides (TMOs) host a wealth of exotic phenomena ranging from charge, orbital, and magnetic order to nontrivial topological phases and superconductivity. In order to translate these unique materials properties into novel device functionalities, TMOs must be doped. However, the nature of carriers in doped oxides and their conduction mechanism at the atomic scale remain unclear. Recent angle-resolved photoelectron spectroscopy (ARPES) investigations provided new insight into these questions, revealing that the carriers of prototypical metal oxides undergo a transition from a polaronic liquid to a Fermi liquid regime with increasing doping. Here, by performing ab initio many-body calculations of the ARPES spectra of TiO$_2$, we show that this transition originates from non-adiabatic polar electron-phonon coupling, and occurs when the frequency of plasma oscillations exceeds that of longitudinal-optical phonons. This finding suggests that a universal mechanism may underlie polaron formation in TMOs, and provides a new paradigm for engineering emergent properties in quantum matter.

Elucidating the nature of charge carriers in doped transition metal oxides is key to understanding the mechanism of electrical conduction in these multifunctional materials. In conducting oxides the infrared-active vibrations can couple strongly to electrons, leading to the formation of polarons. Polarons are electrons dressed by a phonon cloud, and represent a paradigmatic example of emergent state in condensed matter. Depending on their mass and size, polarons exhibit widely different conduction mechanisms, from band-like transport to thermally-activated hopping transport. Despite being central to the science and technology of oxides, little is known about the properties of polaronic states.

The interest in electron-phonon coupling and polaronic quasiparticles in TMOs has been reinvig-
orated by recent ARPES experiments\textsuperscript{[5-9]}. The signature of polaronic behavior in ARPES spectra is the appearance of satellites below the conduction band, at integer multiples of the optical phonon energy. This is reported in Fig. \textsuperscript{[1]a-b for the paradigmatic case doped anatase TiO\textsubscript{2}}\textsuperscript{5}. These pioneering measurements showed that, by increasing the carrier concentration, polaronic satellites gradually evolve into the photoemission kinks observed in metals and superconductors\textsuperscript{[10]}, see Fig. \textsuperscript{[1]c}. It was proposed that this crossover reflects the evolution of charge carriers from polarons to a Fermi liquid\textsuperscript{[5-9]}. In order to clarify the origin of this transition without making any \textit{a priori} assumption about the underlying mechanism, first principles calculations are urgently called for. However, the investigation of polaronic features in ARPES spectra from first principles and their evolution with doping is exceptionally challenging and has never been reported before.

In the following we focus on the prototypical example of anatase TiO\textsubscript{2}. On top of its well-known applications in solar energy harvesting\textsuperscript{[11,12]} and superhydrophilic technology\textsuperscript{[13,14]}, this material is also being investigated in the quest for transparent conducting oxides based on non-toxic and Earth-abundant elements\textsuperscript{[15,16]}. Despite its pivotal role in a broad range of technologies, the nature of the charge carriers in anatase is still controversial\textsuperscript{17}. Here we address these issues by calculating ARPES spectra and polaron wavefunctions entirely from first principles. We develop a novel theoretical and computational framework that allows us to investigate polarons and Fermi liquid quasiparticles on the same footing, and without resorting to any empirical parameters. Using this new approach, we show how the interplay between the dynamical screening of the electron plasma and the Fröhlich electron-phonon coupling is responsible for the transition between polaronic and Fermi liquid states. We propose that the mechanism identified in this work may be universal, and also applies to other oxides such as SrTiO\textsubscript{3} and ZnO.

**Results**

**Angle-resolved photoemission spectra**

Our calculated ARPES spectra are shown in Fig. \textsuperscript{[1]d-f}, for the same doping levels as in the measurements of ref. \textsuperscript{[5]} reproduced in Fig. \textsuperscript{[1]a-c}. These maps show the bottom of the conduction band of \textit{n}-doped anatase TiO\textsubscript{2}, for three doping levels in the range $10^{18}$ to $10^{20}$ cm\textsuperscript{-3}. All the spectra exhibit a bright parabolic band, whose size increases with doping. This reflects the rise of the Fermi
energy inside the conduction band as the electron density increases. Besides this bright feature, panels a-b (experiments) and d-e (calculations) show each a pair of satellites, a bright one at a binding energy around 0.1 eV, and a dim one near 0.25 eV. These features are identified as polaronic effects. Moving on to higher doping in panels c and f, the satellites disappear and are replaced by band structure kinks near 0.1 eV. Overall, our calculated ARPES spectra are in remarkable agreement with the experiments of ref. [5] In order to achieve this unprecedented level of precision without any adjustable parameters, we developed an innovative computational framework.

In our calculations the photoelectron intensity maps are obtained using the single-particle spectral function $A(k, \omega)$, where $\hbar k$ and $\hbar \omega$ are the electron momentum and binding energy, respectively, and $\hbar$ is the reduced Planck constant. The spectral function is calculated using the state-of-the-art cumulant expansion, which has been employed recently to successfully describe plasmon satellites. The cumulant expansion can naturally be applied to investigate the spectral properties of a polaronic system since the theory stems from the exact solution of the Fröhlich electron-boson coupling Hamiltonian. In this formalism the spectral function is expressed as:

$$A(k, \omega) = \frac{1}{2\pi} \sum_n \text{Re} \int_{-\infty}^{+\infty} dt e^{i(\omega - \varepsilon_{nk}/\hbar)t} C_{nk}(t),$$

where $n$ is the electron band index, $\varepsilon_{nk}$ is the electron eigenvalue in absence of many-body interactions, and $t$ is the time variable. The quantity $C_{nk}$ is the so-called ‘cumulant’ function, and can be calculated by using the standard electron-phonon self-energy $\Sigma_{nk}$ as a seed (see Methods and Supplementary Note 1):

$$\Sigma_{nk}(\omega) = \frac{1}{\hbar} \sum_{mn\nu} \int_{\Omega_{BZ}} d\mathbf{q} \left| g_{mn\nu}(k, \mathbf{q}) \right|^2 \left[ \frac{n_{\mathbf{q} + \mathbf{k} + \mathbf{q}} + f_{\mathbf{q} + \mathbf{k} + \mathbf{q}}}{\omega - \varepsilon_{\mathbf{q} + \mathbf{k} + \mathbf{q}}/\hbar + \omega_{\mathbf{q} + \mathbf{k} + \mathbf{q}} - i\eta} + \frac{n_{\mathbf{q} + \mathbf{k} + \mathbf{q}} + 1 - f_{\mathbf{q} + \mathbf{k} + \mathbf{q}}}{\omega - \varepsilon_{\mathbf{q} + \mathbf{k} + \mathbf{q}}/\hbar - \omega_{\mathbf{q} + \mathbf{k} + \mathbf{q}} - i\eta} \right].$$

Here $g_{mn\nu}(k, \mathbf{q})$ is the electron-phonon vertex, and describes the probability amplitude for an electron in the initial state $|nk\rangle$ to be scattered into the final state $|mk + q\rangle$ by a phonon with momentum $\hbar \mathbf{q}$ and energy $\hbar \omega_{\mathbf{q} + \mathbf{k}}$ in the branch $\nu$. The terms $n_{\mathbf{q} + \mathbf{k} + \mathbf{q}}$ and $f_{\mathbf{q} + \mathbf{k} + \mathbf{q}}$ denote the Bose-Einstein and Fermi-Dirac occupations, respectively; $\Omega_{BZ}$ is the Brillouin zone volume, and $\eta$ a positive infinitesimal. The numerical evaluation of Eqs. (1)-(2) is very challenging owing to the singular nature of the Fröhlich interaction at long wavelengths. To overcome this challenge we use the Wannier function technique of refs. [28][29], as implemented in the EPW code [30].
Figure 1:  | *Ab initio* ARPES spectra of n-doped anatase TiO$_2$. (a-c) ARPES spectra of anatase TiO$_2$ measured by Moser et al.$^5$ The measurements were taken at 20 K on samples with doping concentrations $5 \times 10^{18}$ cm$^{-3}$ (a), $3 \times 10^{19}$ cm$^{-3}$ (b), and $3.5 \times 10^{20}$ cm$^{-3}$ (c). The zero of the energy is set to the Fermi level. The electron momentum $k_x$ is along the $\Gamma \Sigma$ line of the anatase Brillouin zone (see panel j). Reproduced with permission from ref.[5] copyright 2013 American Physical Society. (d-f) Calculated spectral function of anatase TiO$_2$, for the same electron momenta and nominal doping levels as in a-c (indicated in each panel in units of cm$^{-3}$). Gaussian masks of widths 25 meV and 0.015 Å$^{-1}$ were applied to account for the experimental resolution.$^5$ (g-i) Band structures extracted from the calculated spectral functions in d-f. The bare bands are in red, the bands including electron-phonon interactions are in blue. The calculated mass enhancement parameter $\lambda$ is indicated in each panel. (j) Brillouin zone and high-symmetry lines of anatase TiO$_2$. (k) Calculated ARPES spectrum for a doping concentration of $3 \times 10^{19}$ cm$^{-3}$, showing the anisotropy of the electron dispersions along $\Gamma X$ (basal plane of the tetragonal lattice, see Supplementary Fig. 1) and $\Gamma Z$ ($c$-axis).

The strength of the electron-phonon interaction is most commonly expressed in terms of a single parameter $\lambda$, which describes the enhancement of the electron mass from the band effective mass $m_b$ to the polaron mass $m^*$ via $m^* = m_b(1 + \lambda)$, as well as the renormalization of the Fermi velocity$^{31}$. To determine this parameter we first extract the band structures underlying Fig.[1]d-f, by tracking the maxima in the energy distribution curves. This procedure yields a set of parabolic...
bands and their satellites in Fig. 1g-h, and a distorted parabola in Fig. 1i (blue curves). For comparison we also report the electronic bands in absence of electron-phonon interactions (red curves). For carrier concentrations of $5 \times 10^{18}$ and $3 \times 10^{19}$ cm$^{-3}$, polaron satellites are clearly visible in Fig. 1g-h, whereas at $3.5 \times 10^{20}$ cm$^{-3}$ we see a band structure kink but no satellites. From these band structures we obtain the mass enhancement parameter as the ratio between the Fermi velocity of the bare band, $v^F_0$, and that of the dressed band, $v^F_F$, as indicated in Fig. 1i: $\lambda_k = v^F_0/v^F_F - 1$, where we explicitly included the dependence on the wavevector at the Fermi surface. As we move from the lowest to the highest doping level we obtain $\lambda_k = 0.73, 0.70, \text{and } 0.20$, respectively. Our calculated value at intermediate doping is in excellent agreement with the mass enhancement determined in experiments, $\lambda = 0.7$. 

In Fig. 1k we show the spectrum calculated at intermediate doping for electron momenta along $\Gamma X$ and $\Gamma Z$, as well as the Fermi surface cut. Here we see that the Fermi surface pocket is elongated along the $\Gamma Z$ direction. This elongation reflects the anisotropic character of the band effective masses, which we calculate to be $m^\perp = 0.40 m_e$ and $m^\parallel = 4.03 m_e$ along $\Gamma X$ and $\Gamma Z$, respectively ($m_e$ is the electron mass). Surprisingly this anisotropy is not reflected in the electron-phonon coupling strength: our calculations indicate that the mass enhancement $\lambda_k$ varies by less than 10% along the [100], [110], and [001] directions, leading to an average value of $\lambda = 0.73, 0.70$ and 0.19 for the three doping levels considered (see also Supplementary Note 2). These results are in good agreement with resonant inelastic X-ray scattering experiments on anatase TiO$_2$. 

**Origin of satellites and kinks**

Having established that our calculations can accurately reproduce experimental spectra without adjustable parameters, we now proceed to identify the mechanisms that drive the formation of polaron satellites by selectively turning off individual components of the calculations. The energy separations between the quasiparticle bands and the first satellite in Fig. 1h and Fig. 1e are 106 meV and 124 meV, respectively. In the case of Fig. 1f the kink appears at a binding energy of 100 meV. These energy scales are compatible with a Fröhlich-type coupling to the longitudinal-optical (LO) $E_u$ phonon at 109 meV (see phonon dispersions in Supplementary Fig. 1). This vibrational mode corresponds to the stretching of the Ti-O bonds in the $ab$ plane, as shown in Fig. 2e. Another candidate bosonic mode is the $c$-axis $A_{2u}$ phonon at 88 meV, however the energy of this mode appears
too small to account for the satellites and kinks in Fig. 1. In order to quantify the importance of the $E_u$ phonon, in Fig. 2a we compare two calculations: the complete spectrum at intermediate doping (left) and a calculation where the coupling to modes with energy above 100 meV is artificially suppressed (right). We see that, upon removing high-energy phonons, the intensity of the first satellite decreases and the second satellite disappears. The effective mass is also visibly lower, in fact the analysis of the mass enhancement parameter yields $\lambda = 0.3$, to be compared to the total coupling $\lambda = 0.7$. It follows that the $E_u$ phonon contributes $\sim 60\%$ of the total coupling, hence it represents the primary mechanism behind the satellites.

In Fig 2b we test the importance of many-body correlations beyond the one-shot Migdal approximation. On the left we show the spectrum at intermediate doping, obtained from the cumulant expansion; on the right we show the result of a calculation within the one-shot Migdal approximation (see Methods). The Migdal approximation is obtained from the complete electron-phonon self-energy by neglecting the three-point vertex. In this approximation the electron-phonon self-energy contains only non-crossing diagrams. The additional approximation adopted in first principles calculations is to replace the fully renormalized electron and phonon propagators by those evaluated within density functional theory. We name this choice the ‘one-shot’ Migdal approximation to emphasize the lack of self-consistency in the electron propagator. From Fig. 2b we notice that the one-shot Migdal approximation fails twice: firstly, the separation between the quasiparticle band and the first satellite is too large as compared to experiment (151 meV instead of $\sim 100$ meV); secondly, the dim satellite around 0.25 eV is completely missing. This test highlights the crucial role of high-order electron-phonon correlations in the description of polarons in conducting oxides. It would be interesting to see whether the deficiencies of the one-shot Migdal approximation could be avoided by performing a fully self-consistent Migdal calculation, and to establish the importance of crossing diagrams which are included in the cumulant expansion; however this test is currently out of reach.

We now move to discuss the origin of the crossover between satellites and kinks in the spectra as a function of doping. The Fröhlich theory of polarons considers a single electron added to a polar insulator. This description does not take into account that, as more electrons are added to the system, the polar electron-phonon coupling is weakened by the electronic screening of the
Figure 2: **Origin of satellites and kinks, and polaron wavefunctions in anatase TiO$_2$.** (a) Effect of high-energy phonons: we compare the spectral function calculated by taking into account all vibrational modes (left half-panel, for $3 \times 10^{19}$ cm$^{-3}$) and a calculation where all phonons with energy above 100 meV have been eliminated (right half-panel). (b) Effect of electron-phonon correlations beyond the one-shot Migdal approximation: the left half-panel shows a complete calculation using the cumulant expansion method, while in the right half-panel the spectral function is calculated within the one-shot Migdal approximation. The doping level is the same as in a. (c) Effect of dynamical screening on the spectral function: the left half-panel shows the spectral function calculated for the highest doping level ($3.5 \times 10^{20}$ cm$^{-3}$) by taking into account the screening of the electron-phonon interaction by carriers. In the right half-panel this effect is turned off, and as a result the electron-phonon coupling is artificially enhanced. For clarity these spectra were not convoluted with Gaussian masks as in Fig. 1d-f. (d) Identification of polaronic region and Fermi liquid region in $n$-doped anatase TiO$_2$: the red spheres represent the plasmon energy at each doping level, the horizontal line is the energy of the LO $E_u$ phonon. The electron-phonon coupling strength $\lambda$ is given by the blue spheres. The lines are guides to the eye. (e) Ball-and-stick representation of the LO $E_u$ phonon, showing for clarity only one of the TiO$_6$ octahedra. (f) Square moduli of the polaron wavefunctions near the origin and further away from the origin, in the polaronic region (blue, $3 \times 10^{19}$ cm$^{-3}$) and in the fermi liquid region (gold, $3.5 \times 10^{20}$ cm$^{-3}$). The corresponding envelope functions are shown in the bottom half of the figure as the blue and red curves, respectively. These wavefunctions are extended over all three Cartesian directions, but are only shown along the $c$-axis for clarity.
charge carriers. Following ref. [2], we treat this issue by screening the electron-phonon vertex by the frequency-dependent Lindhard dielectric function with the calculated effective mass and dielectric permittivity of anatase TiO$_2$ (see also Methods). For completeness we show in Supplementary Fig. 2 the dielectric screening as a function of carrier density and how the Fröhlich electron-phonon vertex is influenced by doping. In order to illustrate the importance of carrier screening, here in Fig. 2c we compare two scenarios: the spectrum calculated at high doping by accounting for electronic screening (left); the same system, but this time ignoring the screening of the electron-phonon coupling by doped carriers (right). The result is striking: in the absence of screening one obtains a sharp polaron satellite, in stark disagreement with experimental evidence. This comparison indicates that a correct description of the electronic screening is absolutely crucial to capture the photoemission kink at high doping. On the contrary, when we repeat the comparison of Fig. 2c for the cases of low and intermediate doping, we find a completely different picture: at these doping levels the electronic screening does not play any significant role.

These conflicting observations can be rationalized by inspecting the timescales of lattice vibrations and electronic screening. The $E_u$ phonon vibrates with a period $T_{\text{ph}} = 38$ fs. The characteristic response time of the electronic screening is set by the plasma frequency of doped carriers, $T_{\text{el}} = 2\pi/\omega_p$. In the case of $n$-doped TiO$_2$ the electrons occupy a singly-degenerate parabolic band minimum, therefore $\omega_p = (ne^2/\epsilon_0\epsilon_\infty m_b)^{1/2}$, where $n$ is the electron density, $\epsilon_0$ the permittivity of vacuum, and $\epsilon_\infty$ the high-frequency dielectric constant of TiO$_2$. For the electron densities considered in Fig. 1a-c we calculate $T_{\text{el}} = 122$ fs, 50 fs, and 15 fs, respectively. From these values we deduce that, at the lowest doping, the carriers are too slow to screen the long-range electric field generated by the oscillation of the $E_u$ phonon (122 fs vs. 38 fs). In this case the screening is ineffective and the Fröhlich interaction dominates the spectrum. On the contrary, at the highest doping the electrons oscillate faster than the LO phonon (15 fs vs. 38 fs). In this case the electronic screening is almost complete and the Fröhlich coupling is largely suppressed. In this regime the strength of the kink depends critically on the carrier concentration; with increasing doping the coupling to the LO phonons is gradually suppressed, and the ARPES spectrum is dominated by the weaker coupling of carriers to non-polar phonons [8]. These considerations are summarized in Fig. 2d, where we compare the energy of the $E_u$ phonon with the plasma energy $\hbar\omega_p$ of the carriers, and we monitor the evolution of the coupling strength with doping. We can identify two
regions: a polaronic regime, corresponding to the situation $\omega_p < \omega_{ph}$, and a Fermi liquid regime, corresponding to $\omega_p > \omega_{ph}$. In the polaronic region the electronic screening is ineffective, we see satellites in the spectra, and the electron-phonon mass enhancement is not sensitive to the doping level. In the Fermi liquid region the Fröhlich coupling is strongly suppressed, polaron satellites are gradually replaced by photoemission kinks, and the coupling strength decreases. To further validate this trend, we performed additional calculations for a doping concentration of $1 \times 10^{20}$ cm$^{-3}$ in the transition region. We found that one satellite is still present in the spectrum (Supplementary Fig. 3). A careful investigation of the mass renormalization parameter yields $\lambda = 0.34$, thus confirming that the electron-phonon coupling is weakened by the electronic screening. The present analysis reveals that the origin of the crossover from polarons to a Fermi liquid in the ARPES spectra of doped TiO$_2$ is to be found in a novel form of electron-phonon coupling, which we refer to as a ‘non-adiabatic Fröhlich interaction’. The non-adiabatic treatment of the electronic screening is especially important to correctly capture the response of the electrons at low doping.

Given the qualitative change in the band structures at the crossover from the polaronic to the Fermi liquid regime, it is natural to ask how this evolution is reflected in the wavefunctions. In order to explore this aspect we calculate the polaron wavefunctions by generalizing the perturbation theory of ref. 2 to ab initio calculations. The top of Fig. 2f shows the square moduli of the polaron wavefunctions at the bottom of the conduction band near the maximum of the polaron wavefunction and a few unit cells away: on the left the intermediate doping level, which is inside the polaronic region of Fig. 2d; on the right the high doping level, in the Fermi liquid region. The bottom half of Fig. 2f shows the corresponding envelope functions. Here we see that the wavefunction of an electron in the Fermi liquid region is essentially akin to a periodic Bloch function. On the contrary, the wavefunction of an electron in the polaronic region shows spatial localization. To quantify the ‘size’ of the polaron in this latter case we define a polaron radius $r_p$ using the half width at half maximum. From Fig. 2f we obtain $r_p = 5.7$ nm. This result indicates that, despite the qualitative changes in the ARPES spectra, we are in the presence of large polarons throughout the entire doping range, and that polarons in TiO$_2$ are considerably more delocalized than previously thought. Since thermally-activated hopping transport corresponds to $r_p$ of the order of the lattice constant, our finding supports the notion that electrical conduction in anatase TiO$_2$ takes place via standard band-like transport. To avoid ambiguities we emphasize that the present result refers to intrinsic
mobile polarons, not to localized electronic defect states such as those associated with O vacancies and which are not mobile\cite{35}.

**Discussion**

The non-adiabatic Fröhlich mechanism identified here is simple enough that it is likely to play a role in many other conducting oxides. In order to test this hypothesis we estimate the critical density for the crossover in doped SrTiO$_3$\cite{8} and doped ZnO\cite{9}. In SrTiO$_3$ the plasma energy for an electron concentration of $10^{20}$ cm$^{-3}$ is $\hbar \omega_p \sim 64$ meV\cite{36}, therefore to match the 100 meV LO phonon of SrTiO$_3$ one would need a doping level of $2.6 \times 10^{20}$ cm$^{-3}$. In the experiments of ref.\cite{8} the carriers are confined in a thin surface layer corresponding to approximately 3 unit cells, therefore we estimate the critical carrier density for the two-dimensional electron liquid in the range $3 \times 10^{13}$ cm$^{-2}$. This value is remarkably close to the critical density determined experimentally, $4 \times 10^{13}$ cm$^{-2}$\cite{8}. More accurate calculations will need to take into account the two-dimensional screening at the surface of SrTiO$_3$ and its quantum paraelectric nature. A similar analysis can be carried out for ZnO\cite{9}. In this case the energy of the highest LO phonon is 72 meV, while the plasma energy for a surface electron concentration of $7.5 \times 10^{13}$ cm$^{-2}$ is 320 meV\cite{37}. The transition would then appear around $3.8 \times 10^{12}$ cm$^{-2}$, slightly below the density reported in ref.\cite{9}. In agreement with our estimate, the spectra of ref.\cite{9} exhibit a behavior which is intermediate between kinks and satellites.

In summary, our findings indicate that the electron-phonon coupling in TiO$_2$ is more complex than previously thought, and that the non-adiabatic Fröhlich coupling could be the unifying mechanism behind the transition from polarons to Fermi liquids in conducting oxides; explicit *ab initio* calculations will be required to confirm this point. Looking further ahead, our work suggests that a fine control of the interplay between lattice vibrations and plasma oscillations may offer a pathway for investigating novel emergent states in quantum materials, and provide opportunities in the development of quantum technologies based on TMOs.
Methods

Ground-state calculations

*Ab initio* calculations were carried out for anatase TiO₂ (space group *I*4₁/amd), using the experimental lattice parameter \( a = 3.784 \text{ Å}^{33} \). We used density-functional theory (DFT) within the generalized gradient approximation of Perdew, Burke, and Ernzerhof\(^{39}\). The core-valence interaction was described by means of norm-conserving pseudopotentials, with the semicore Ti-3s and Ti-3p states explicitly taken into account. Electron wavefunctions were expanded in a planewaves basis set with kinetic energy cutoff of 200 Ry, and the Brillouin zone was sampled using a \( 6 \times 6 \times 6 \) Monkhorst-Pack mesh. Lattice-dynamical properties were calculated using density functional perturbation theory (DFPT). All DFT and DFPT calculations were performed using the Quantum ESPRESSO\(^{40}\) package.

Electron-phonon coupling

Calculations of electron-phonon couplings were performed using the *EPW* code\(^{30}\), the cumulant expansion was performed separately (see Supplementary Note 1). The Fröhlich electron-phonon matrix element was calculated using the method of ref.\(^{29}\) An accurate description of the Fröhlich vertex as described in ref.\(^{29}\) is essential: ignoring this effect leads to a severe underestimation of the mass enhancement\(^{41}\). To evaluate Eq. (2) we computed electronic and vibrational states as well as the scattering matrix elements on a \( 4 \times 4 \times 4 \) Brillouin-zone grid. These quantities were interpolated with *ab initio* accuracy onto a fine grid with \( 2 \cdot 10^6 \) random \( q \)-points using *EPW*. The positive infinitesimal \( \eta \) was set to 10 meV. Temperature effects were accounted for by including the Fermi-Dirac occupation in the spectral function, corresponding to the experimental temperature of 20 K.

Doping

Doping was included using the rigid-band approximation since the system is degenerate. In fact, by considering the Mott criterion for the metal-insulator transition\(^{42}\), \( a_0^* n_c^{1/3} \approx 0.25 \) where \( a_0^* = \hbar^2 \epsilon_0 / (e^2 m_b) \) is the effective Bohr radius, we obtain a value \( n_c = 1.3 \times 10^{18} \text{ cm}^{-3} \) for the critical density that is below the doping levels investigated. The screening of the electron-phonon interaction arising from the doped carriers was taken into account by computing the dielectric function.
\( \epsilon(q, \omega) \) in the random-phase approximation\(^{2,43} \) for a homogeneous electron gas with the calculated effective mass \( m_b \) and dielectric permittivity \( \epsilon_\infty \) of anatase TiO\(_2\)\(^{2,43} \) (see Supplementary Note 3 for a rationale). In this expression the plasma frequency directly reflects the carrier density, therefore the influence of doping on the dielectric screening in anatase TiO\(_2\)\(^{44} \) is included in the calculations. The resulting non-adiabatic matrix element is \( g_{mn\nu}(k, q) = g_{mn\nu}(k, q) / \epsilon(q, \omega_q + i/\tau_{nk}) \), where \( \tau_{nk} \) is the electron lifetime near the band edge. Here we use \( \hbar/\tau_{nk} = 55 \) meV from ref.\(^5 \). We checked that this choice does not affect our conclusions (Supplementary Fig. 4). We note that, owing to the strong anisotropy of the Fermi surface in \( n \)-doped anatase TiO\(_2\), it is possible that the nominal doping levels estimated in ref.\(^5 \) using the Fermi momentum may underestimate the actual carrier densities in the measured samples.

**Angle-resolved photoemission spectra**

The cumulant function used in the calculation of the ARPES spectra is obtained from the self-energy of Eq. (2) as follows: \( C_{nk}(t) = (\pi \hbar)^{-1} \int_{-\infty}^{\infty} d\omega \theta(\omega - \epsilon_{nk}/\hbar) e^{(i\omega + \eta)t} \text{Im} \Sigma_{nk}(\epsilon_{nk}/\hbar - \omega)/(\omega - i\eta)^2 \), where \( \theta \) is the Heaviside function. More details on the cumulant expansion are provided in Supplementary Note 1. The calculations within the one-shot Migdal approximation shown in Fig. 2b were performed by using directly Eq. (2) inside the spectral function:\(^{26} \) \( A(k, \omega) = 2\pi^{-1} \sum_n |\text{Im} (\hbar\omega - \epsilon_{nk} - \Sigma_{nk})^{-1}|. \) The spectra were computed without including photon matrix elements effects\(^{10} \). This approximation is justified since the variation of the orbital character over the \( \sim 0.1 \) eV binding energy range shown in Fig. 1 is negligible\(^{10} \). In order to extract the bands shown in Fig. 1g-i we proceeded as follows. For the quasiparticle bands, which are the brightest features in Fig. 1d-f, we calculate the poles of the electron Green’s function as \( E_{nk} = \epsilon_{nk} + \text{Re} \Sigma_{nk}(E_{nk}) \), where \( \epsilon_{nk} \) and \( E_{nk} \) are the bare and the renormalized dispersions, respectively. For the satellites in Fig. 1d-e the previous procedure is not applicable since these features are not poles of the Green’s function. In this case we directly identify the maxima in the energy dispersion curves\(^{10} \), and obtain the pairs of parabolic bands which are visible in Fig. 1g-h at binding energies below 0.1 eV.

**Polaron wavefunction**

In order to calculate the wavefunction of the polaron we generalized the perturbation theory approach of ref.\(^2 \) to the case of multiple electron bands and phonon branches, as well as \textit{ab initio}
electron-phonon matrix elements. We found \( \tilde{\psi}_{nk}(r; \{ \tau_{\kappa p} \}) = C \psi_{nk}(r) \times \left[ 1 + \sum_\nu \int \frac{dq}{V_BZ} |g_{nn\nu}(k, q)|^2 e^{i\mathbf{q}\cdot\mathbf{r}/(\varepsilon_{nk} - \varepsilon_{nk+q} - \hbar\omega_{\nu})^2}|0_p\rangle \right] \), where \( \psi_{nk} \) and \( \tilde{\psi}_{nk} \) are the electron wavefunction without and with electron-phonon interactions, respectively. \( \{ \tau_{\kappa p} \} \) indicate the nuclear coordinates, \( |0_p\rangle \) is the phonon ground state and \( C \) is a normalization constant. The details of the derivation are given in Supplementary Note 4. The wavefunctions in Fig. 2 were obtained by setting \( nk \) to the conduction band edge; the envelope functions were calculated by retaining only the term inside the square brackets in the above expression. A random grid with \( 2 \cdot 10^6 \) points was used for the Brillouin-zone integration, and a small imaginary part \( \eta = 10 \text{ meV} \) was added to the denominator.

Acknowledgements

The authors gratefully acknowledge Marco Grioni for granting permission to reproduce Fig. 1a-c, and M. Grioni, B. Gumhalter and P. D. C. King for fruitful discussions. This work was supported by the Leverhulme Trust (Grant RL-2012-001), the Graphene Flagship (Horizon 2020 Grant No. 696656 – GrapheneCore1), and the UK Engineering and Physical Sciences Research Council (Grant No. EP/M020517/1). This work used the ARCHER UK National Supercomputing Service via the AMSEC Leadership project, and the Advanced Research Computing facility of the University of Oxford.

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Origin of the crossover from polarons to Fermi liquids in transition metal oxides

Supplementary Information

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Supplementary Figure 1 | Crystal structure and phonon dispersions of anatase TiO$_2$.  
(a) Ball-and-stick model of the tetragonal unit cell of anatase TiO$_2$. Ti atoms are in gray and O atoms are in red. The lattice vectors and crystallographic directions referred to in the main text are indicated. (b) Calculated phonon dispersion relations of anatase TiO$_2$ along high-symmetry directions of the Brillouin zone. The $E_u$ and $A_{2u}$ phonons discussed in the main text are indicated. The $A_{2u}$ phonon is infrared active only along the $c$ axis, accordingly it is found to give a smaller contribution to the total Fröhlich coupling after integrating over all phonon momenta.
Supplementary Figure 2 | Non-adiabatic electron-phonon matrix elements in anatase TiO$_2$.

(a) Lindhard dielectric function associated with doped carriers in anatase TiO$_2$ at different values of $q$. For illustration we consider the highest doping level, $3.5 \times 10^{20}$ cm$^{-3}$. (b) Non-adiabatic electron-phonon matrix elements $g_{mn\nu}^{NA}(k, q)$ corresponding to the LO E$_u$ phonon of anatase TiO$_2$, as a function of doping level. We set $n_k$ to the bottom of the conduction band. The electron-phonon matrix element becomes weaker at high doping. (c) Comparison between the density of states (DOS) near the conduction band bottom calculated from first principles (red) and modelled with a parabolic band (blue): $DOS(E) = V/(2\pi^2) (2m_b/\hbar^2)^{3/2} \sqrt{E}$, where $V$ is the unit cell volume of anatase. The ab initio DOS was computed using Wannier interpolation to obtain the electronic energies on a randomly generated dense $k$ grid and with a Gaussian smearing of 5 meV. The gray vertical line indicates the energy corresponding to the Fermi level at the highest doping. The parabolic DOS starts deviating significantly from the calculated one for energies above 0.2 eV.
Supplementary Figure 3 | Spectral function in the transition regime. (a) Spectral function of anatase TiO$_2$ calculated for the doping level $1 \times 10^{20}$ cm$^{-3}$. Gaussian masks of widths 25 meV and 0.015 Å$^{-1}$ were applied as in Fig. 1 of the main text. (b) Band structure extracted from a (blue lines) together with the bare band (red line). For this doping one satellite is still visible but the mass renormalization parameter is decreasing ($\lambda = 0.34$ as reported in the main text).
Supplementary Figure 4 | Impact of the electron lifetime on the spectral properties. Spectral function of anatase TiO$_2$ calculated for the doping level $3.5 \times 10^{20}$ cm$^{-3}$ using $\hbar/\tau = 25$ (a), 55 (b) and 75 meV (c) to compute the electronic screening (see Methods). The spectral features are virtually unchanged. We recomputed the mass renormalization parameter $\lambda$ as in the main text which gives $\lambda = 0.19, 0.20$ and 0.22 for increasing broadening. The same analysis for the doping concentration $5 \times 10^{18}$ cm$^{-3}$ yields $\lambda = 0.77, 0.73$ and 0.72 for the same broadenings. These values fall within 10% of the results presented in the main text.
The spectral function is related to the imaginary part of the one-electron retarded Green’s function by:

\[
A(k, \omega) = - \frac{1}{\pi} \sum_n \text{Im} \, G_{nk}(\omega).
\] (1)

In the cumulant expansion the Green’s function is obtained in the time domain, starting from the interaction picture:\n
\[
G_{nk}(t) = i \exp \left[ -i\varepsilon_{nk} t / \hbar + C_{nk}(t) \right],
\] (2)

where \( C_{nk}(t) \) is the cumulant function. By taking the Fourier transform of this expression to the frequency domain and inserting it in Supplementary Eq. (1) we obtain Eq. (1) of the main text.

In order to obtain an expression for the cumulant which is amenable to computation, it is customary to expand the exponential in powers of the cumulant:

\[
G_{nk} = G_{nk}^0 \left[ 1 + C_{nk} + C_{nk}^2 / 2 + \cdots \right],
\]

where \( G_{nk}^0(t) = i \exp (-i\varepsilon_{nk} t / \hbar) \) is the Green’s function in absence of electron-phonon interactions. Alternatively, the Green’s function can be obtained from the Dyson equation:

\[
G_{nk} = G_{nk}^0 + G_{nk}^0 \Sigma_{nk} G_{nk}^0 + G_{nk}^0 \Sigma_{nk} G_{nk}^0 \Sigma_{nk} G_{nk}^0 + \cdots.
\]

By comparing these expansions term-by-term one finds an explicit expression for the cumulant function, which is given in the Methods and is reproduced here for completeness:

\[
C_{nk}(t) = \frac{1}{\pi \hbar} \int_{-\infty}^{\infty} d\omega \frac{\text{Im} \, \Sigma_{nk}(\varepsilon_{nk} / \hbar - \omega)}{(\omega - i\eta)^2} e^{(i\omega + \eta)t} \theta(\varepsilon_F - \varepsilon_{nk} + \hbar\omega).
\] (3)

In practical calculations the self-energy \( \Sigma_{nk} \) is replaced by the best available approximation, which is the Migdal expression given in Eq. (2) of the main text. A rigorous derivation of the cumulant expansion and a discussion of its advantages and limitations can be found in Supplementary Refs. 4,5.

In particular, in Supplementary Ref. 4 it is shown that the choice of seeding the self-energy calculated with the first non-crossing diagram also guarantees that no overcounting of correlated higher-order contributions is introduced in the theory. The inclusion of crossing diagrams in the evaluation of the Green’s function results from the time orderings of the \( t \) variables in the cumulant expansion.

The spectral function given in Eq. (1) of the main text yields multiple bosonic satellites, one for each term in the Taylor expansion of \( \exp[C_{nk}(t)] \). A convenient expression for the case of a
single satellite was derived in Supplementary Ref. 3. In this work we extended the method of Supplementary Ref. 3 to the case of multiple satellites. Following Supplementary Ref. 3, we write the spectral function as:

\[
A(k, \omega) = \sum_n \left[ A_{nk}^{QP}(\omega) + A_{nk}^{QP}(\omega) \ast A_{nk}^{S1}(\omega) + A_{nk}^{QP}(\omega) \ast A_{nk}^{S2}(\omega) + \cdots \right],
\]  

(4)

where \(\ast\) indicates the convolution. In the last expression the quasiparticle contribution \(A_{nk}^{QP}(\omega)\) is defined as:

\[
A_{nk}^{QP}(\omega) = \frac{2}{\pi} \frac{|\text{Im} \Sigma_{nk}(\epsilon_{nk})|}{[\hbar \omega - \epsilon_{nk} - \text{Re} \Sigma_{nk}(\epsilon_{nk})]^2 + [\text{Im} \Sigma_{nk}(\epsilon_{nk})]^2},
\]

(5)

and the satellite contributions associated with one-phonon and two-phonon processes are:

\[
A_{nk}^{S1}(\omega) = \int_{-\infty}^{\infty} dt \ e^{i \omega t} C_{nk}(t), \quad A_{nk}^{S2}(\omega) = \int_{-\infty}^{\infty} dt \ e^{i \omega t} \frac{1}{2} C_{nk}(t) C_{nk}(t).
\]

(6)

The function \(A_{nk}^{S1}\) can be written in terms of the electron-phonon self-energy by combining Supplementary Eqs. (6) and (3) and carrying out the Fourier transform. This step was performed in Supplementary Ref. 3:

\[
A_{nk}^{S1}(\omega) = \frac{\beta_{nk}(\omega) - \beta_{nk}(\epsilon_{nk}) - (\omega - \epsilon_{nk}/\hbar) \frac{\partial \beta_{nk}}{\partial \omega} |_{\epsilon_{nk}/\hbar}}{(\hbar \omega - \epsilon_{nk})^2},
\]

(7)

with

\[
\beta_{nk}(\omega) = \frac{1}{\pi} \text{Im} \Sigma_{nk}(\epsilon_{nk}/\hbar - \omega) \theta(\epsilon_F/\hbar - \omega).
\]

(8)

In order to obtain the contribution of the second satellite, we note that \(A_{nk}^{S1}(\omega)\) is the Fourier transform of \(C_{nk}(t)\), therefore the expression for \(A_{nk}^{S2}(\omega)\) in Supplementary Eq. (6) can be rewritten using the convolution theorem:

\[
A_{nk}^{S2} = \frac{1}{2} A_{nk}^{S1} \ast A_{nk}^{S1}.
\]

(9)

By combining Supplementary Eqs. (9) and (4) we obtain the expression used in our calculations:

\[
A(k, \omega) = \sum_n \left[ A_{nk}^{QP}(\omega) + A_{nk}^{S1}(\omega) \ast A_{nk}^{QP}(\omega) + \frac{1}{2} A_{nk}^{S1}(\omega) \ast A_{nk}^{S1}(\omega) \ast A_{nk}^{QP}(\omega) + \cdots \right].
\]

(10)

From Supplementary Eq. (10) we can extrapolate the general expression for the case of many satellites:

\[
A(k, \omega) = \sum_n \left[ 1 + A_{nk}^{S1}(\omega) \ast \frac{1}{2} A_{nk}^{S1}(\omega) \ast A_{nk}^{S1}(\omega) + \cdots \right] A_{nk}^{QP}(\omega) = \sum_n e^{A_{nk}^{S1}(\omega) \ast A_{nk}^{QP}(\omega)}.
\]

(11)
When considering a drastically simplified model system consisting of dispersionless electrons and an Einstein phonon spectrum, this last expression reduces to the well-known Lang-Firsov series of polaron satellites. In fact, by setting $A^{QP} = Z\delta(\omega)$ and $A^{S1} = \lambda\delta(\omega - \omega_{ph})$, Supplementary Eq. (11) gives $A(\omega) = Z\sum_{m=0}^{\infty}(\lambda^m / m!) \delta(\omega - m\omega_{ph})$. By further requiring the normalization of the spectral function, that is $\int A(\omega) d\omega = 1$, we obtain $Z = e^{-\lambda}$ and the Lang-Firsov expression is recovered. In our \textit{ab initio} calculations we truncate Supplementary Eq. (11) to the second order. In practice we first evaluate the quasiparticle and satellite contributions, and then we perform two successive numerical convolutions in order to obtain the two satellites which are seen in the experiments.

**Supplementary Note 2 | Mass enhancement and Fröhlich coupling constant**

The mass enhancement parameters obtained from the quasiparticle bands along the $\Gamma\Sigma$, $\Gamma X$, and $\Gamma Z$ directions are $\lambda = 0.73, 0.73$, and 0.74 at the lowest doping; $\lambda = 0.70$ in each direction at the intermediate doping; and $\lambda = 0.20, 0.20$, and 0.18 at the highest doping. The mass enhancement can also be calculated directly from the energy derivative of the real part of the electron self-energy at the Fermi level, $\lambda_k = -\partial \text{Re} \Sigma_k(\omega) / \partial \omega|_{\omega=\epsilon_F}$. We checked that the values thus obtained fall within less than 10% of the ones listed above, the average over the Fermi surface being $\lambda = 0.68, 0.65$ and 0.19 for the three values of doping considered. This approach also validates our calculations of the contribution to $\lambda$ arising from different phonon modes presented in the analysis of Fig. 2a in the main text.

From the mass enhancement parameter it is also possible to obtain the quasiparticle strength $Z$ as $Z = 1/(1 + \lambda)$. Using the values of $\lambda$ reported in Fig. 1g-i we obtain $Z = 0.58, 0.59$, and 0.83 with increasing doping. Our calculated quasiparticle strength at intermediate doping overestimates the experimentally-determined value, $Z = 0.36$. We attribute this difference to extrinsic losses not accounted for in our calculations; these losses are known to transfer spectral weight to the satellites.

In polar materials it is customary to describe the coupling to a dispersionless LO phonon via the
dimensionless Fröhlich coupling constant $\alpha$:

$$\alpha = \frac{e^2}{\hbar} \left( \frac{m_b}{2 \hbar \omega_{LO}} \right)^{1/2} \left( \frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0} \right),$$  \hspace{1cm} (12)

where $m_b$ is the band mass, $\hbar \omega_{LO}$ the energy of the LO phonon, $\epsilon_0$ and $\epsilon_\infty$ the static and high-frequency permittivities, respectively. We evaluate Supplementary Eq. (12) using the experimental values of $\omega_{LO}$, $\epsilon_0$, and $\epsilon_\infty$ from Supplementary Ref. 11. For the effective masses we use our DFT calculations since no accurate experimental values are available: $m_\perp = 0.40 m_e$ and $m_\parallel = 4.03 m_e$. By using these parameters in Supplementary Eq. (12) we find the experimental Fröhlich constants $\alpha_\perp = 1.0$ and $\alpha_\parallel = 3.4$, and their isotropic average $\alpha = 1.8$. The Fröhlich constant $\alpha$ can also be obtained from $\textit{ab initio}$ calculations of the electron-phonon matrix elements, following Supplementary Ref. 12. In this case we calculate $\alpha_\perp = 0.9$, $\alpha_\parallel = 3.0$, and the isotropic average $\alpha = 1.6$. These values are in excellent agreement with experiment, therefore our description of Fröhlich coupling in anatase TiO$_2$ is expected to be highly accurate. We note that the anisotropy of the Fröhlich coupling constant $\alpha_\perp$ and $\alpha_\parallel$ does not stem from anisotropic electron-phonon interactions, but rather from the strong anisotropy in the band masses, which enter $\alpha$ as seen in Supplementary Eq. (12).

For weak and intermediate couplings, the polaron effective mass is commonly estimated using $m^* = m_b (1 - \alpha/6)^{-1}$ \cite{13,14}. While this procedure is adequate for isotropic crystals, it cannot be used in the present case of TiO$_2$. In fact, if we use the isotropic average of the coupling constant, then we underestimate $m^*$ with respect to experiment\cite{8}. On the other hand, if we consider $\alpha_\perp$ and $\alpha_\parallel$ separately, then we obtain a strongly anisotropic mass enhancement, which is not consistent with our many-body calculations. These observations indicate that, in the case of anisotropic crystals, the Fröhlich constant $\alpha$ should be used with caution.

**Supplementary Note 3 | Dielectric screening**

In this work we calculate the additional screening arising from the charge carriers using the random phase approximation (RPA) for the homogeneous electron gas, that is the Lindhard screening. This choice is motivated by the fact that we must evaluate the screening for millions of electron-phonon matrix elements, and explicit $\textit{ab initio}$ calculations of the RPA screening for such a dense Brillouin
zone grid are not currently feasible. The use of the Lindhard model is justified by the fact that the
system under consideration lies in the high-density electron-gas limit. To confirm this point we
evaluate the Wigner-Seitz radius $r_s$ given by $r_s = (4\pi n a_0^3/3)^{-1/3}$, where $n$ is the doping density
and $a_0^* = h^2\varepsilon_0/(e^2m_b)^{15}$. Using the band effective mass $m_b$ and the static dielectric constant $\varepsilon_0$ of
anatase we obtain $r_s = 1.6$ for the lowest doping level $5 \times 10^{18}$ cm$^{-3}$. This value is comparable
or even smaller than what found in simple metals, therefore the use of the electron gas model to
describe doped carriers is justified. Moreover, the electronic bands are to a good approximation
parabolic in the energy range considered in this work; this is seen by comparing the ab initio
density of states with the parabolic band model, Supplementary Fig. 2c. Therefore the use of a
Lindhard function is justified, and we expect this choice to be very accurate in the present case.

**Supplementary Note 4 | Polaron wavefunction**

In order to calculate the wavefunction of a polaron, we follow the approach of Supplementary
Ref. 15 and express the many-body electron-phonon state using Rayleigh-Schrödinger perturbation
theory. We consider a system at zero temperature and with a single electron added to the bottom
of the conduction band. The resulting expression is:

$$
\tilde{\psi}_{nk}(r; \{\tau_{\kappa}\}) = \psi_{nk}(r) |0_p\rangle + \frac{1}{\sqrt{N_q}} \sum_{m\nu q} g_{mn\nu}(k, q) \psi_{mk+q}(r) \hat{a}_{-q\nu}^\dagger |0_p\rangle,
$$

(13)

where $\{\tau_{\kappa}\}$ are the nuclear coordinates, $\hat{a}_{-q\nu}^\dagger$ is a phonon creation operator, and $|0_p\rangle$ is the ground
state with no phonons. The Brillouin zone is discretized using a uniform grid with $N_q$ phonon
wavevectors. Since the atomic displacements are smaller than characteristic interatomic distances,
we can simplify the above expression by replacing $\hat{a}_{-q\nu}^\dagger$ (which is a function of the normal mode
coordinates) by its average over a given electron-phonon state$^{15}$. In order to determine a lower
bound to the polaron radius, we evaluate this expectation value by considering an electronic wave-
function localized at the center of the reference frame, $r = 0$. The result is:

$$
\langle \hat{a}_{-q\nu}^\dagger \rangle = \frac{1}{\sqrt{N_q}} \sum_{m'k'} g_{m'nk\nu}(k, q) \frac{\varepsilon_{mk} - \varepsilon_{mk+q} - \omega_{q\nu}}{\varepsilon_{nk} - \varepsilon_{mk} - \omega_{q\nu}}.
$$

(14)
By replacing $\langle \hat{a}_-^{\dagger} \rangle$ inside Supplementary Eq. (13) we have:

$$
\tilde{\psi}_{nk}(r; \{\tau_\kappa\}) = \frac{e^{ik\cdot r}}{\sqrt{\Omega}} \left[ u_{nk}(r) + \frac{1}{N_q} \sum_{mnq} g_{mn\nu}(\kappa, \kappa) u_{mk+q}(r) e^{iq\cdot r} \sum_{m'} \frac{g^{*}_{m'n\nu}(\kappa, \kappa)}{\varepsilon_{nk} - \varepsilon_{mk+q} - \omega_{\kappa\nu}} \delta_{mm'} \right] |0_p\rangle. 
$$

(15)

The expression for $\tilde{\psi}_{nk}$ given in the Methods was obtained from Supplementary Eq. (15) by considering the normalization of the wavefunction and by retaining the long-wavelength part of the Fröhlich vertex, so that $g_{mn\nu}(\kappa, \kappa) \rightarrow g_{mn\nu}(\kappa, \kappa) \delta_{mn}$. We already demonstrated that this approximation is very accurate for anatase TiO$_2$. Since the main contribution to Supplementary Eq. (13) arises from long-wavelength optical phonons, we also replace $u_{mk+q}(r)$ by $u_{mk}(r)$, in the spirit of the $k \cdot p$ approximation.

Our choice of calculating the polaron wavefunction within Rayleigh-Schrödinger perturbation theory is justified since this theory is valid in the range $\alpha \leq 5$ (see Supplementary Ref. 15, p. 516), and the largest Fröhlich coupling constant in anatase TiO$_2$ is $\alpha_\parallel = 3.0$ (see Supplementary Note 2).

We note that the present many-body approach for calculating the polaron wavefunction differs significantly from DFT calculations of excess electrons in oxides using large supercells. In fact in the present case we employ a quantum description of both electrons and nuclei, and a dynamical (non-adiabatic) description of their interactions. In contrast, supercell DFT calculations describe the nuclei as classical particles and decouple electronic and nuclear degrees of freedom using the Born-Oppenheimer adiabatic approximation. These approximations are not justified in the case of TiO$_2$, and have led to results which are very sensitive to the DFT exchange and correlation functional and the size of the supercell. Our present perturbative approach does not suffer from these shortcomings.
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