Oscillator-strength ratio of two Fe XVII soft X-ray transitions essential for plasma diagnostics finally agrees with theory

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One of the most enduring and intensively studied problems of X-ray astronomy is the disagreement of state-of-the-art theory and observations for the intensity ratio of two ubiquitous Fe XVII transitions of crucial value for plasma diagnostics, dubbed 3C and 3D. We unravel this conundrum.

Soft X-ray spectra from present space observatories such as Chandra [1] and XMM-Newton [2, 3] offer deep insight into massively energetic astrophysical sources, and together with steadily improving future missions such as XRISM [4] and Athena [5, 6] will continue improving our understanding of phenomena driving the growth of galaxies, stellar formation, and the reionization phase of the universe. Interpretation of their data has to rely on accurate atomic-structure calculations gained from experimentally benchmarked theory methods. With ubiquitous iron ions being the strongest emitters in most X-ray sources, decades-long efforts have aimed at understanding their spectra, with a plethora of papers delivering valuable diagnostics on a multitude of sources [7–13]. However, a long-lasting discrepancy in a key result, namely the intensity ratio $I_{3C}/I_{3D}$ of the strong resonance line 3C (\(2p^6\)\(J=0\) \(\rightarrow\) \(\{2p^6\}_{1/2}\) \(3d_{3/2}\)\(J=1\)) and the intercombination line 3D (\(2p^6\)\(J=0\) \(\rightarrow\) \(\{2p^6\}_{3/2}\) \(3d_{5/2}\)\(J=1\)) in the dominant ionic species of many plasmas, Fe XVII (Fe\(^{16+}\)), repeatedly raised questions on the accuracy of calculations [14, 15].

Since Fe XVII has a Ne-like closed-shell configuration, describing its single excitations was expected to be easier than for any ions with semi-filled $L$ or $M$ shells. This disagreement raised questions over atomic-structure predictions in general, and remained until today a major concern in the field [16].

Early space and laboratory soft-X-ray spectrometers with resolving powers of $E/\Delta E \leq 1000$ could not separate from 3D the satellite line C of Fe XVII (\(2p^63s\)\(J=1/2\) \(\rightarrow\) \(\{2p^5\}_{1/2}\) \(3s3d_{5/2}\)\(J=3/2\)), resulting in erroneous values of $I_{3C}/I_{3D}$ well below 3.0. After this was understood, measurements in more pure Fe XVII plasmas gave slightly larger values of $\approx 3.0$ [18, 19]. Predictions based on distorted-wave and R-matrix theory using small sets of configurations typically yielded values of $\approx 4.0$ and above [20–23]. With the gradual inclusion of more configurations and relativistic effects, predictions converged towards $I_{3C}/I_{3D} \approx 3.5$ [24–26].

Tentative explanations hinged on a key point: Most experiments used electron-impact excitation, and thus cascades from higher states, resonant excitation, po-
FIG. 1. Fluorescence yield of the soft X-ray transitions 3C and 3D of Fe XVII as well as B and C of Fe XVI as a function of the exciting-photon energy. Fitted Voigt profiles (orange) show residuals (bottom panel) due to a non-perfectly Gaussian distribution of the monochromator spectral bandwidth. Inset: Comparison of this (blue) with the LCLS measurement (green [15]) and a Capella observation (red [17]).

Ionization, a non-Maxwellian electron-energy distribution, population transfer, or other effects could be the cause [27, 28]. It was also suggested that the measured $I_{3C}/I_{3D}$ ratio could differ from the theoretical oscillator-strength ratio $R = f_{3C}/f_{3D}$. Indeed, some related predictions seemed to be closer to observations [29, 30]. Meanwhile, the free-electron laser Linac Coherent Light Source (LCLS) enabled resonant photoexcitation experiments directly probing the oscillator-strength ratio and not affected by electron-impact excitation [15], but the result of $R = 2.61 \pm 0.21$ departed from theory even more than earlier works. Non-linear excitation dynamics [31–33] and charge-state population transfer [34] were suggested as the cause. Renewed experimental efforts with a synchrotron-radiation-based technique [35] brought much better resolution than the LCLS data, and excluded population transfer and non-linearities. The result $R = 3.09 \pm 0.1$ [35] still disagreed with the much-improved calculations from the same work which considered all known quantum mechanical effects, predicting $R = 3.55 \pm 0.05$. This value was confirmed by additional numerous large-scale calculations using different codes. To solve this, we tried to resolve the natural linewidths of 3C and 3D, both well below 20 meV.

In this Letter, we present new measurements of 3C and 3D in Fe XVII ions applying soft X-ray resonant photoexcitation with an increase in both the resolving power by a factor of 2.5 and the signal-to-noise ratio (SNR) by three orders of magnitude, as well as improved calculations that reduce the theoretical uncertainty by a factor of 2.5, all finally leading to mutual agreement.

The experiment was performed with an electron beam ion trap (EBIT) [36], PolarX-EBIT, at the beamline P04 of PETRA III. EBIT operation at synchrotron-radiation facilities for providing highly charged ions (HCI) has been described elsewhere [35, 37–42]. EBITs use an electron-beam energy $E_B$ higher than the respective ionization threshold of the HCI species of interest, here $IP_{Fe^{16+}} \approx 490$ eV. Since electron-impact ionization cross sections start at zero at threshold and peak at energies two to three times higher than it, we chose $E_B \approx 1200$ eV. However, this causes a very strong background by non-resonant electron-impact excitation, which we suppressed by time-coincident detection in experiments using pulsed excitation at free-electron lasers [15, 43, 44]. There, the stochastic nature of self-amplified spontaneous emission pulses can lead to non-linear excitation, a systematic effect that we originally underestimated in Ref. [15].

Synchrotron-radiation sources have pulse-peak intensities many orders of magnitude lower than free-electron lasers, but at much higher repetition rates. In our recent work on $R$ at PETRA III [35], the photon-bunch separation of 192 ns hindered us in measuring energy-resolved fluorescence in time coincidence. Hence, the SNR was only $\approx 0.05$, since fluorescence after photoexcitation was much weaker than the background due to the high electron-beam energy used for producing enough Fe XVII. Nonetheless, since in Ref. [35] the excitation spectra reached an eight times higher resolution than in Ref. [15], the total uncertainty of $R$ was substantially reduced.

For reduction of that strong background, we cycled
\(E_B\) within tens of us (see Fig. 2 in the Supplemental Material [45]) between a high value for breeding Fe XVII fast enough \((E_B = 1200 \text{ eV})\) and a lower one of \(\approx E_B = 250 \text{ eV}\) for recording the fluorescence, well below the threshold for electron-impact excitation, and avoiding discrete energies where resonant and non-resonant photorecombination also cause strong background. By optimizing both duty cycle and slew rate for maximum resonant fluorescence signal, we achieved a SNR \(\approx 50\), roughly thousand times better than in our former work [35]. We could choose a monochromator-exit slit width of 25 μm and keep a strong signal. We optimized the monochromator at the branch 1 of the P04 beamline until reaching its highest specified spectral resolution [46].

A possible alternative method for background suppression, the magnetic trapping mode [47], proved here less efficient due to the low magnetic field of PolarX-EBIT [48].

We measured 3C and 3D of Fe XVII as well as two lines from Fe XVI, named B \([(2p^63s)_{j=1/2} \rightarrow (2p^5)_{1/2}(3s3d)_{3/2}j=1/2]\) and C \([(2p^63s)_{j=1/2} \rightarrow (2p^5)_{1/2}(3s3d)_{3/2}j=3/2]\) with a resolving power of \(E/\Delta E = 20000\) (see Fig. 1) and excellent SNR of \(\approx 50\). This unprecedented data quality made it possible to reliably fit to each transition a Voigt profile, with a Gaussian component due to Doppler broadening and monochromator resolution, and a clearly separable Lorentzian component.

The spectra unveil the hitherto hidden Lorentzian wings, a possibility already considered in [49]. Blends are the main problem at low resolution. When it becomes good enough to separate C from 3D, unmodelled line wings hidden in the background (as in our work [35]) still alter the fitted intensities (see Fig. 6). At the even higher, present resolution we can unambiguously distinguish these wings from a linear background, allowing us to fit statistically significant Voigt profiles and extract relative intensities with uncertainties below 1%.

Crucially, integrated intensities become nearly free from blends, clarifying why the old discrepancy had apparently persisted in our recent work [35]. The results also have smaller systematic uncertainties from background fluctuations or photon-flux variations.

Our experimental result is \(R = 3.51 \pm 0.02_{\text{stat}} \pm 0.07_{\text{sys}}\). Due to its accuracy, it supersedes all reported experimental results. Moreover, it agrees well with our present large-scale calculations and other very recent state-of-the-art works (see Fig. 3 and Table 1 in the Supplemental Material [45]).

We have noticed an additional systematic effect in a later campaign at beamline P04 on a different target. While scanning the photon energy for recording the EBIT fluorescence, we let the photon beam pass through it and reach the electron spectrometer ASPHERE III [50] installed downstream, and simultaneously monitored with very high relative resolution the excess energy of copious photoelectrons emitted from a clean gold surface. Unexpected oscillations revealed that the angular encoder measuring the grating rotation applied an obsolete interpolation table causing a periodic compression and stretching of the monochromator energy axis by \(\approx \pm 0.04 \text{ eV}\) with \(\approx 1 \text{ eV}\) period (see Supplemental Material [45]). We modelled how this distorts the Voigt profiles finding a systematic uncertainty from this source of \(\approx 2\%\), which now dominates the final error budget. Another effect is a modulation of the measured linewidths that affects our determinations (widths \(\approx 25 \text{ meV}\)) at the level of 10%.

In general, the oscillator-strength ratio of two transitions with upper states decaying only to their respective initial lower state, as in the case of 3C and 3D, is correlated with the corresponding ratio of natural linewidths: \(\Gamma_{3C}/\Gamma_{3D} = R \cdot (E_{3C}/E_{3D})^2\). Hence, by combining the observed \(R = 3.51 \pm 0.09\) with the energy quotient \((E_{3C}/E_{3D})^2 = 1.032\) determined in earlier experiments [62], a linewidth ratio \(\Gamma_{3C}/\Gamma_{3D}\) of \(\approx 3.6\) is expected. In
contrast, the present ratio of the fitted Lorentzian widths of 3C and 3D lies well below 2.0. We found the reason by measuring the Lorentzian widths of well-known He-like transitions $K_\alpha$, $K_\beta$, and $K_\gamma$ of F VIII, for which theory is very accurate. This revealed a pseudo-Lorentzian instrumental contribution that adds to their natural linewidths. It arises from beamline diffraction and mimics at finite resolution a Lorentzian profile (J. Viefhaus, R. Follath, private communication) [63]. Since this spurious effect is nearly constant within the photon-energy range $E_{3C} - E_{3D} \approx 13.4$ eV, we could extract the difference $\Gamma_{3C} - \Gamma_{3D} = 10.92(175)$ meV. By combining this value with the measured $R$, we obtained the natural linewidths (see Table V). Analogously, for lines B and C we added their respective difference from the 3D Lorentzian width to the natural linewidth of 3D determined here. The influence of the periodic energy shifts of the monochromator on the linewidth were also modelled, giving a relative systematic uncertainty of 15%, the largest in the final error budget.

We performed a large-scale configuration-interaction (CI) calculation for Fe XVII (see Supplemental Material [45] for details) with a new version of our highly scalable parallel CI code, where we substantially enlarged the basis in relation to our previous calculations [35] in order to tackle the last remaining parameter that affected the energy calculations. The new basis includes all orbitals up to 24spdf/g12h, versus only up to 12sp17df/g in [35], and we allow for the same excitations of initial configurations for the larger basis when constructing the set of CI configurations as in that work. We find a very small change (53-60 meV) of the level energies, demonstrating a high level of convergence. The $3^1D_1$ and $1^1P_1$ energies agree with the experiment [64] to 0.01%. The effect on $R$ is negligible, $\Delta R \approx 0.001$. With this, we have saturated the CI expansion, given that the configuration-function space in Ref. [35] already included triple and other excitations and opening of the 1s$^2$ shell. The present computation includes over 1.2 million configurations and almost 100 million Slater determinants, with the Breit contribution taken into account. We included QED effects in the computations following Refs. [35, 65] by using several different QED potentials described in Ref. [65]. This choice caused only negligible differences on the energies and oscillator-strength ratio. While QED contributes $-0.016$ to the ratio, mostly through its effect on the electric-dipole matrix elements, the QED relative influence on the 3C and 3D energies is negligible. In a conservative way, we consider the total QED correction as uncertainty of our final prediction of $R = 3.55 \pm 0.02$. The calculated transition rates for 3C and 3D transitions, including small contributions from other transitions of 0.07% and 0.23%, are $2.239(2) \times 10^{13}$ s$^{-1}$ and $6.12(2) \times 10^{12}$ s$^{-1}$, and the corresponding natural linewidths are 14.74(1) meV and 4.03(1) meV. We calculated the difference of the 3C and 3D linewidths to be $13.44(2)$ meV, with the QED contribution of 0.02 meV that is much larger than the uncertainty in the electronic correlation. The QED effects for the linewidth difference arise from subtle differences in configuration mixing for the upper and lower states of 3C and 3D. It would be also very interesting to measure the energy differences between $2s^22p^33s$ and $2s^22p^53d$, where the QED contribution is five to ten times larger and thus easier to probe (see Supplemental Material [45]), while

![FIG. 3. Comparison between experimental (red colored vertical bars including 1-sigma uncertainty) and theoretical results of this work as well as selected theoretical and experimental literature values for the 3C/3D oscillator-strength ratio $R$ (white - experiment). Theoretical approaches [20, 21, 23–27, 32, 34, 35, 51–57] are marked as follows: Distorted wave (magenta hexagons), multi-configuration Dirac-Fock (teal diamonds), R-Matrix (blue triangles), many-body perturbation theory (orange crosses), and configuration interaction (black circles). Blue band: observed line ratios in astrophysical sources [7, 8, 58–61], with color shades coding the distribution of values weighted by their reported accuracies. Figure based on Table III from Supplemental Material [45].]

| Line          | Experiment (meV) | Theory (meV) |
|---------------|------------------|--------------|
| 3C - 3D       | 10.92(175)       | 10.71(2)     |
| 3C            | 15.27(247)       | 14.74(1)     |
| 3D            | 4.22(68)         | 4.028(15)    |
| B             | 16.42(301)       | 14.43‡       |
| C             | 20.52(380)       | 23.10‡       |
all electronic correlation effects are of comparable size. We find that the uncertainty of the QED calculations is the dominant one in the predictions of the 3C-3D splitting and their oscillator-strength ratio. A further reduction of experimental uncertainties will be thus crucial, and offers a unique opportunity to benchmark QED effects for both energies and natural linewidths in sufficiently complicated 10-electron systems with all other theoretical influences controlled to high accuracy. Such tests are particularly important for the development of HCl-based optical clocks [66], since experimental searches for clock transitions require theory predictions including QED effects that can be computed with our code.

After forty years of experimental and theoretical efforts, we have found a satisfactory explanation for the long-standing emission problem in Fe XVII. The new evidence from both experiment and theory now strengthens confidence in input from state-of-the-art theory to advanced astrophysical models and the resulting interpretations of various observations. Moreover, our work exposes how critical a good understanding of non-Gaussian lineshapes in both theory and experiment is for the determination of transition amplitudes and line positions, especially when resolving power, as in currently operating missions, is limited such that line wings mimic a linear background [67].

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SUPPLEMENTAL MATERIAL

The following sections contain Supplemental Material on technical details of the experiment, as well as extended tables from the data analysis and theory calculations.

EXPERIMENT

Setup

We show the general scheme of the experiment in Fig. 4. PolarX-EBIT is installed in the first focal position behind the monochromator exit port at branch 1 of the PETRA III beamline P04. Its position is mechanically adjusted to ensure the best possible overlap with the narrow photon beam in increments of $\approx 20 \mu m$, and daily checked. At the trap center, the highly charged ions are confined within a cylindrical volume of 18 mm length and $\approx 200 \mu m$ diameter, which the photon beam passes along the trap axis. Two windowless silicon-drift detectors with 80 and 150 mm$^2$ sensitive areas, respectively, are mounted side-on at mutually orthogonal directions.

Electron-beam energy switching cycle

In our last measurements [35], the Fe XVII target was produced by an electron beam at an energy of approximately 1600 eV. For the present measurement campaign, the measurement scheme uses a cycle including both an ion-breeding and a probing phase, see Fig. 5. During the ion-breeding phase, a sufficient amount of Fe XVII was produced by the electron beam at energies close to the values employed during the first measurement campaign. After this phase, the electron-beam energy was reduced to only 260 eV within a few ms for mitigating the background induced by both electron-impact excitation and photorecombination. In this probing phase, the electron beam does not have enough energy to produce any undesired strong background signal in the photon-energy region around 800 eV. The beam energy during the probing phase was selected such that dielectronic-recombination resonances, which have cross sections orders of magnitude larger than the direct radiative recombination were avoided as much as possible. However, the ions still experience radiative and charge-exchange recombination. Due to these constant losses and the lack of any production channels for Fe XVII, the population of these ions is slowly depleted during the probing phase. After the Fe XVII population has almost entirely recombined during the probing phase, the cycle started over, and new ions were bred at the higher electron-beam energies; usually, hundreds of ms were required to produce enough Fe XVII. The lifetime of Fe XVII in the probing phase was usually in the order of tens to hundreds of ms, depending on several parameters such as electron-beam current, Fe(CO)$_5$ injection pressure, and residual gas pressure, as well as on the excitation of auto-ionizing transitions driven by the photon beam. Using the so-called magnetic-trapping mode, i. e., completely turning off the electron beam and radically confining the ion cloud purely by the magnetic field as demonstrated in [68, 69], was unfeasible for such long times in the PolarX-EBIT, since its magnetic field strength is several times lower than in superconducting EBITs, thus the radial drift of the ion orbits increases too fast. Utilizing the breeding-probing scheme, the laser-induced fluorescence could be observed almost entirely background-free and thus, the signal-to-noise ratio could be improved by three orders of magnitude compared to the previous beamtime.

Measurement stability

We acquired the data presented in this paper within 16 hours, primarily during a night shift. Within this shift, no instabilities such as electron-beam current variations or beam dumps in the storage ring PETRA III, or Fe(CO)$_5$ injection-pressure variations in the PolarX EBIT were recorded. This experimental stability is reflected by constant amplitudes observed for both lines of Fe XVII 3C and 3D as well as B and C of Fe XVI, see left panel of Fig. 6. In the right panel of Fig. 6, the individual 3C/3D oscillator-strength ratio of 16 consecutive measurements as well as their weighted average (red) is depicted, resulting in a ratio of 3.51 with a statistical uncertainty of only 0.57%. The same stability is also found in the observed Lorentzian widths of all four lines, see Fig. 7, resulting in a statistical uncertainty ranging between 1% for 3C and 5% for 3D.

Systematic uncertainties

Region-of-interest selection

One of the largest systematical uncertainties in our measurements presented in Ref. [35] arose from a poor signal-to-noise ratio (SNR) resulting in large variations of the final result depending on the selected region-of-interest (ROI) used to project the two-dimensional data to spectra. In this work, the SNR is by far sufficient to clearly separate the photon-beam-induced fluorescence signal from the electron-impact-induced background, see Fig. 8, completely eliminating any possible systematic uncertainties due to the ROI selection.
Background and photon-flux instabilities

In contrast to our previous measurements performed at P04, during this campaign a photodiode registering the incident photon-beam flux was mounted downstream of the experimental setup (see Fig. 4). Hence, the photon flux irradiating the trapped ion cloud was simultaneously monitored, and the observed line intensities accordingly corrected. Beside the expected 1% flux variation due to the top-up filling mode of the storage ring PETRA III and slight efficiency differences of the beamline due to energy-dependent X-ray-optic reflectively, no anomalies were observed.

The stability of the ion target production was continuously tracked by monitoring the electron-beam current, the Fe(CO)$_3$ injection pressure, and the low energy background below 500 eV.

Detection efficiency

In front of both commercial windowless detectors used (Ketek Vitus H80 & Ketek Vitus H150), a 500 nm thick aluminum filter was employed to prevent visible light from saturating them. Since 3C and 3D have different energies, the filter transmission changes for each of them. We corrected the observed intensities based on literature values for the respective transmissions [70]. The thickness tolerance of the aluminum filter was specified within ±10%. This results in an uncertainty of the transmission correction based on the literature values for 450 nm and 550 nm thick filters of approximately 0.13%. We assume a constant detector efficiency across the small energy range between 810 and 830 eV.

Non-linear effects

In order to explain the unexpectedly low value of the 3C/3D oscillator-strength ratio measured at LCLS, non-linear effects due to the high photon-flux peak intensity of the FEL were proposed [33, 71]. The radiative lifetimes of 3C and 3D are predicted to be 163 fs and 45 fs, respectively, and are thus in the same order of magnitude as the X-ray pulse lengths of LCLS of the experiment in Ref. [15] between 200 fs and 2000 fs [33]. It was hence proposed that the high flux of the free-electron laser was sufficient to populate the upper level of 3C and 3D at different rates within these short laser pulse. According to Oreshkina et al. [71], a peak flux density of at least $1 \times 10^{12} \text{W cm}^{-2}$ or more is required to observe such non-linear effects during the measurement. The photon peak intensity at the synchrotron beamline P04 is estimated based on the number of photons registered by the cali-
FIG. 6. Left panel: Observed amplitudes for 3C and 3D as a function of the measurement time. Right panel: Inferred 3C/3D amplitude ratio for two consecutive scans at a time. The red solid and dashed lines indicate the weighted average and statistical uncertainty.

FIG. 7. Observed Lorentzian linewidths of the Voigt profiles applied to the measured fluorescence of the lines 3C (blue), 3D (orange), B (green), and C (red). The solid lines represent the weighted average of all measured linewidths of a given transition including the 1-σ deviation (dashed lines).

brated diode downstream of the experimental setup

\[\Psi_{\text{Photonbeam}} = 4 \times 10^{11} \text{ photons/s.} \quad (1)\]

Given the employed monochromator energy of

\[E = 825 \text{ eV} = 1.32 \times 10^{-16} \text{ J}, \quad (2)\]

an average power of

\[P_{\text{average}} = 5.28 \times 10^{-5} \text{ W} \quad (3)\]

is obtained. During the measurements, PETRA III operated in timing mode, resulting in a photon-bunch repetition rate of 5.21 \(\times\) 10\(^6\) pulses/s. Combined with a minimal possible focal spot size of 1 \(\times\) 10\(^{-10}\) m\(^2\) and a typical photon-bunch length of 44 ps, a peak flux density of

\[\rho \approx 2.3 \times 10^9 \text{ W m}^{-2} \quad (4)\]

is obtained, which is orders of magnitude below the required value predicted by theories. It is interesting to note that the separation between two photon pulses of 192 ns was sufficiently long for the excited states of 3C and 3D, with lifetimes of tens to hundreds of fs, to decay to their ground states before the next photon pulse arrived. At the estimated photon peak flux and the long photon-pulse separation, non-linear effects of any kind, even for the most conservative assumptions of a minimal focus spot size and minimal bunch lengths, can be explicitly excluded for the measurements presented within this work.
After the ions were bred, the electron-beam energy was lowered to a fraction of the upper value to suppress background. Thereby, the lower electron-beam energy of 265 eV was no longer sufficient to produce the investigated charge states Fe XVI and Fe XVII. Due to recombination caused by interaction with the electron beam (radiative recombination) as well as with neutral residual gas (charge exchange), the population of highly charged ions was continuously depleted. The recombination rates of both charge states Fe XVII and Fe XVI were expected to be similar, but since Fe XVII recombined into Fe XVI, the latter was continuously fed resulting in an increasing Fe XVI/Fe XVII abundance ratio during the probing cycle obtained, as seen by comparing the areas of the lines C and 3D as a function of time, see Fig. 9. Since Fe XVII was the highest possible charge state produced during the breeding period, Fe XVII could not be produced by recombination from other, higher charge states.

In the LCLS measurements, the resolving power was insufficient to separate line 3D from C. Hence, it was proposed that the strong Auger-Meitner decay channel of the upper state of C to the ground state of 3D in Fe XVII combined with the high photon-flux intensity of the FEL caused a so-called population-transfer mechanism, i.e., a change of the plasma charge-state distribution during the measurements.

We investigated this effect in this work. The time evolution of the 3C/3D intensity ratio after switching down the electron-beam energy is depicted in Fig. 9. Even though the relative abundance of Fe XVI almost doubled during one probing cycle, the 3C/3D oscillator-strength ratio remained stable. To verify that the observed 3C/3D oscillator-strength ratio was constant and thus independent of the Fe XVI/Fe XVII abundance ratio, a linear model was fitted to the oscillator-strength ratio evolution. As expected, the fit agreed with a constant value. Hence, the 3C/3D oscillator-strength ratio was independent of the Fe XVI/Fe XVII abundance ratio, and charge-state population-transfer mechanisms are explicitly excluded.

Angular-encoder interpolation error

During an independent campaign performed at the PETRA III beamline P04 some months after the measurements presented here, we found that the angular encoder used to determine the grating rotation did not perform as expected. Unfortunately, we found an oscillation distorting the monochromator photon-energy axis. The photon energy of the plane mirror, plane grating monochromator (PGM) at beamline P04 is calculated with the angles of the mirror and the grating measured by their corresponding rotary encoders. The encoder glass scales are segmented at regular intervals of 0.01 degrees, which relates, e.g., to ≈7 eV for the grating and ≈4.5 eV for the mirror (around 800 eV, fix focus constant \( cff = 3.4 \)). A 14-bit interpolation is applied on the quadrature signals of each encoder. In principle, four photodiodes measuring the intensity transmitted between optical masks imprinted on the encoder disks capture periodically oscillating signals with different phases. Transforming these patterns into angular increments involves a careful and accurate intensity interpolation and normalization of the signal amplitudes of the different diodes. Measurements using the photoelectron spectrometer ASPHERE III [50] located at P04 revealed periodic and reproducible departures with an amplitude of ±40 meV from the otherwise
FIG. 9. Observed 3C/3D oscillator-strength ratio (blue, left axis) and the Fe XVI/Fe XVII abundance ratio (orange, right axis) as a function of time after switching down the electron beam. The red solid line shows the result of a linear function fitted to the oscillator-strength ratio values. Dashed lines indicate the 1-σ uncertainty.

FIG. 10. Orange: measured difference between the actual and the demanded monochromator energy utilizing the ASPHERE III photoelectron spectrometer [50]. Blue: The acquired dataset is modeled and extrapolated using a combination of four sinusoidal waveforms.

(over large angular displacements) linear energy scale (see Fig. 10 of this Supplemental Material). A study of such angular calibration effects at the Swiss Light Source [72] showed that the interpolation errors typically did not interfere with measurements up to monochromator energy resolutions of $E/\Delta E \approx 10,000$. However, in our present measurement, the effect is noticeable, since the encoder specifications of the P04 monochromator are different, and our resolution is twice as high.

The oscillations from Fig. 10 can be well described by a combination of four sinusoidal waveforms with periods of $\approx 1\text{–}2\text{ eV}$ around 800\text{ eV}, and are related to the aforementioned interpolation procedure. Recovering this accordion-like systematic periodic shift is in principle possible using the photoelectron spectrometer, since our statistical uncertainty in the line-centroid determinations is smaller than 1\text{ meV}. Unfortunately, such correction measurements were not performed here. Therefore, the influence of the periodic deviations on the oscillator-strength ratio had to be investigated by simulations. For this purpose, a synthetic periodic deviation of the photon energy consisting of the product of four sinusoidal waveforms was simulated, which in shape, amplitude, and periodicity approximately corresponded to the observed oscillations.

Two synthetic Voigt profiles corresponding to the energies, amplitudes, and linewidths of 3C and 3D, respectively, were generated. Afterwards, each energy for which the intensities of the Voigt profiles were calculated was shifted by the simulated energy deviation. The shifted profiles were then analyzed using the same algorithms used for the analysis of the actual data, and the fit re-
TABLE II. Comparison of 3C/3D oscillator-strength ratio results including error budgets of Ref. [35] and this work.

| Measurement                                                                 | Kühn et al. 2020 [35] | This work |
|----------------------------------------------------------------------------|------------------------|-----------|
| Number of scans (incl. B and C of Fe XVI)                                  | 6+11                   | 60        |
| Resolving power $E/\Delta E$ (FWHM)                                       | 8250                   | 20000     |
| Signal-to-noise ratio                                                     | $\approx 0.05$         | $\approx 45$ |
| Model used                                                                 | Gaussian               | Voigt     |
| 3C/3D oscillator-strength ratio                                           | 3.09                   | 3.51      |
| Statistical uncertainty                                                   | $\pm 2.58\%$          | $\pm 0.57\%$ |
| **Systematical uncertainties**                                            |                        |           |
| ROI selection                                                              | $\pm 1.8\%$           | X         |
| Background instabilities                                                  | $\pm 1.0\%$           | X         |
| Area underestimation of Gaussian profiles fitted to Voigt lines           | possible               | excluded  |
| Detection efficiency uncertainty                                           | $\pm 0.13\%$          | $\pm 0.13\%$ |
| Monochromator interpolation errors                                        | possible               | $\pm 2.0\%$ |
| **Final 3C/3D oscillator-strength ratio**                                 | 3.09(10)               | 3.51(7)   |

FIG. 11. Histogram of simulated relative shifts of the 3C/3D intensity ratio. Two synthetic Voigt lines were disturbed by simulated interpolation errors of the monochromator. The amplitudes of the disturbed lines were analyzed and compared with the initial parameters. Red bars represent the 1-$\sigma$ standard deviation.

The relative change of the amplitude ratio due to simulated interpolation errors is depicted in Fig. 11. We find a distribution of the relative ratio shifts that is similar to a normal distribution centered at 1.0. Hence, on average, the ratio shift induced by incorrect energy interpolations is expected to be negligible. However, the periodic oscillations are assumed to be constant throughout the measurements for similar operating parameters, unless the correction tables of the angular encoders are changed. Hence, measurements of the same energy range will always result in the same oscillating deviation between the actual and the demanded photon energy.

In order to assess this effect, the 1-$\sigma$ standard deviation of the present simulation result of approximately 2% was added to the error budget of the final 3C/3D oscillator-strength ratio value. Using the same simulation program, the influence of this effect on the observed Lorentzian linewidths was also investigated and resulted in systematical uncertainties of up to 15%, completely dominating the error budget of the natural linewidth determinations.

**Error budget**

The systematic and statistical uncertainties, as well as the final error budget of this work and of our previous measurement are listed in Tab. II.
THEORY: CALCULATION OF THE 3C/3D LINE-INTENSITY RATIOS OF Fe\(^{16+}\)

We start from the solution of the Dirac-Hartree-Fock equations in the central-field approximation to construct one-particle orbitals. These calculations are carried out using a configuration-interaction (CI) method, correlating all 10 electrons of the Ne-like ion and taking into account the Breit interaction in all calculations. QED effects are considered following the method outlined in Ref. [65]. During the procedure, basis sets of increasing sizes are used to check for convergence of the values. Each of these is designated by the highest principal quantum number of each partial wave included. As an example, the label \([12spdfg]\) means that all orbitals up to \(n = 12\) are included for the \(spdfg\) partial waves. We compared two different methods of constructing the basis sets and found both leading to the same results. We also noticed that the inclusion of the \(6-12h\) orbitals did not modify the results, and therefore, we omitted higher partial waves.

The CI many-electron wave function \(\Psi_n\) is obtained as a linear combination of all Slater determinants \(\Phi_i\) of a given parity: \(\Psi_n = \sum_i c_i \Phi_i\). The energies and wave functions are determined from the time-independent many-electron Schrödinger equation \(H\Psi_n = E_n\Psi_n\). Contributions to the \(E1\) reduced matrix elements \(D(3D) = D(2p^6 1S_0 - 2p^53d^3 D_1)\) and \(D(3C) = D(2p^6 1S_0 - 2p^53d^1 P_1)\) and the ratio of the respective oscillator strengths

\[
R(3C/3D) = \left( \frac{D(3C)}{D(3D)} \right)^2 \times \frac{\Delta E(3C)}{\Delta E(3D)}
\]

are calculated and listed below.

We start with all possible single and double excitations to any orbital up to \(12spdfg\) from the \(2s^22p^6, 2s^22p^53p, 2s2p^63s, 2s^22p^54f\) even and \(2s^22p^53s, 2s^22p^53d, 2s2p^63p, 2s^22p^54s, 2s^22p^54d\) odd configurations, correlating 8 electrons.

Contributions to the energies of Fe XVII are calculated and listed in Table III. The results are compared with a revised analysis of tabulated experimental data [64]. We use \(LS\) coupling and NIST data term designations for comparisons but note that \(jj\) coupling would be more appropriate for this ion.

Contributions to the \(D(3C)\) and \(D(3D)\) \(E1\) reduced matrix elements and the \(3C/3D\) ratio are listed in Table IV, respectively. The \(3C/3D\) energy ratio is 1.01654. We find \(R = f_{3C}/f_{3D} = 3.55 \pm 0.02\).

We include additional configurations obtained by excitations from the \(1s^2\) shell and to the \(6-12h\) orbitals and list them as “1\(s^2\)” and “12\(h\)” in Tables III and IV. The contributions from the \(1s^2\) shell improve the agreement with the experiment for energies, but have only a very small (-0.006) effect on \(f_{3C}/f_{3D}\). Inclusion of the \(6-12h\) orbitals gives negligible corrections to both energies and matrix elements. Contributions from triple excitations were also found to be negligible in a previous calculation [35], and we did not recalculate them here. We expand the basis set from \([12spdfg]\) to \([17spdfg]\), and then to \([24spdfg]\), and find a modest improvement of the energies compared to the experiment, but a very small shift of \(R\) by -0.003. The last line of Table III shows the difference of the \(3C\) and \(3D\) energies in eV, with the final value being \(3C - 3D = 13.44\) eV.

As an independent test of the quality and completeness of the current basis set, we compare the results for \(D(3C)\) and \(D(3D)\) obtained in length and velocity gauges for the \([12spdfg]\) basis, see rows I and V in Table IV. The difference in the results is only 0.001. Calculations were also done using a completely different B-spline basis set at the level of \([17spdfg]\), with energy differences of no more than 0.03% between the two basis sets, with energies of the B-spline basis set further away from experimental values. The value of the ratio \(R\) differed by 0.0064 between the two basis sets.

We have also compared different QED potentials described in Ref. [65]. All QED corrections in Tables III and IV were calculated with the QEDMOD variant (see Ref. [65]). We found negligible maximum energy differences of -0.7 cm\(^{-1}\), -13 cm\(^{-1}\), and 3 cm\(^{-1}\), between the different potentials. Compared to the semi-empirical approach, there was a maximum energy difference of 94 cm\(^{-1}\). The energy difference was negligible in each comparison.

Transition rates of all other transitions contributing to the radiative decay of the \(3C\) and \(3D\) levels were also calculated. The totals of these rates are small and listed in Table IV. The linewidth value corresponds to the total transition rate.
TABLE III. Contributions to the energies of Fe$^{16+}$ calculated with increased-size basis sets and number of configurations. The results are compared with experiment. All energies are given in cm$^{-1}$ with exception of the last line that shows the difference of the 3C and 3D energies in eV. The basis set is designated by the highest quantum number for each partial wave included. For example, [12spdfg] means that all orbitals up to $n = 12$ are included for spdfg partial waves.

| Configuration | Expt. [64] | [12spdfg] | +1s$^2$ | +[12h] | +[17spdfg] | +[24spdfg] | QED | Final | Diff. [64] | Diff. [64] |
|---------------|-----------|-----------|---------|--------|-----------|-----------|-----|-------|------------|------------|
| 2s$^2$2p$^5$  | $^3S_0$   | 0         | 0       | 0      | 0         | 0         | 0   | 0     | 0          | 0          |
| 2s$^2$2p$^3$3p | $^3S_1$   | 6093295   | 6090490 | 200    | -2        | 933       | 480 | 70    | 6092171    | 1124       |
| 2s$^2$2p$^3$3p | $^3D_2$   | 6121484   | 6118934 | 217    | -5        | 861       | 433 | 56    | 6120496    | 988        |
| 2s$^2$2p$^3$3p | $^3D_3$   | 6134539   | 6131883 | 210    | -4        | 881       | 447 | 107   | 6133524    | 1015       |
| 2s$^2$2p$^3$3p | $^1P_1$   | 6143639   | 6141023 | 218    | -5        | 865       | 432 | 93    | 6142626    | 1013       |

TABLE IV. Contributions to the E1 reduced matrix elements $D(3D) = D(2p^6\,^1S_0 - 2p^33d\,^3D_1)$ and $D(3C) = D(2p^6\,^1S_0 - 2p^33d\,^3P_1)$ (in a.u.) and the ratio of the respective oscillator strengths $R$. See caption of Table III for designations. $L$ and $V$ rows compare results obtained in length and velocity gauges for the [12spdfg] basis. All other results are calculated using the length gauge. Transition rates and linewidth are listed at the bottom of the table. Total of the other transition rates contributing to the lifetime of the 3C and 3D levels are listed in row “Other transitions”.

| Configuration | $D(3C)$ | $\Delta D(3C)$ | $D(3D)$ | $\Delta D(3D)$ | $R(3C/3D)$ | $\Delta R$ |
|---------------|---------|----------------|---------|----------------|------------|------------|
| [12spdfg]     | $L$     | 0.33523        | 0.00000 | 0.17883        | 3.572      | 3.573      |
|               | $V$     | 0.33546        | 0.17893 | 0.00006        | 3.566      | -0.006     |
|               | +1s$^2$ | 0.33505        | -0.00018 | 0.17889        | 0.00000    | 3.566      | -0.006     |
|               | +[12h]  | 0.33523        | 0.00000 | 0.17884        | 0.00001    | 3.572      | 0.000      |
| [17spdfg]     | 0.33522 | -0.00001       | 0.17889 | 0.00006        | 3.570      | -0.002     |
| [24spdfg]     | 0.33520 | -0.00002       | 0.17890 | 0.00001        | 3.569      | -0.001     |
| QED           | 0.33520 | -0.00013       | 0.17890 | 0.00033        | 3.546      | -0.016     |

Recommended transition rate (s$^{-1}$): $2.239(2)\times10^{13}$, $6.11(2)\times10^{12}$
Other transitions (s$^{-1}$): $1.49\times10^{10}$, $1.38\times10^{10}$
Total rate (s$^{-1}$): $2.239(2)\times10^{13}$, $6.12(2)\times10^{12}$
Linewidth (meV): 14.74(1), 4.028(15)
TABLE V. Comparison of the oscillator-strength ratio $f_{3C}/f_{3D}$ and natural linewidths of 3C and 3D between this work, available experimental datasets, and a selection of theoretical predictions. If available, $n_{\text{Config}}$ corresponds the number of included configurations in the calculation. Additionally, the employed method of each measurement or calculation is given: EIE (electron-impact excitation), RSXES (resonant soft-X-ray excitation spectroscopy), DW (distorted wave), RM (R-Matrix), MBPT (many-body perturbation theory), MCDF (multi-configuration Dirac-Fock), CI (configuration interaction), BP-CI (Breit-Pauli configuration interaction). Note that the validity of theory published by Mendoza et al. [55] has been disputed [73]. All linewidths and linewidth differences are given in meV FWHM.

| Method          | $f_{3C}/f_{3D}$  | $\Gamma_{3C} - \Gamma_{3D}$ | $\Gamma_{3C}$  | $\Gamma_{3D}$  | $n_{\text{Config}}$ |
|-----------------|------------------|-----------------------------|----------------|----------------|---------------------|
| **This work**   |                  |                             |                |                |                     |
| RSXES           | 3.51(7)          | 10.92(175)                  | 15.27(247)     | 4.22(68)       |                     |
| **Experiments** |                  |                             |                |                |                     |
| Brown (2001) [74] | EIE             | 1.90(11) - 3.04(12)        |                |                |                     |
| Beiersdorfer (2004) [75] | EIE | 2.04(42) - 3.33(56)        |                |                |                     |
| Brown (2006) [19] | EIE             | 2.98(30)                    |                |                |                     |
| Gillaspy (2011) [29] | EIE | 1.96(14) - 2.78(11)        |                |                |                     |
| Bernitt (2012) [15] | LS             | 2.61(23)                    |                |                |                     |
| Kühn (2020) [35] | LS              | 3.09(10)                    |                |                |                     |
| **Astrophysical observations** |               |                             |                |                |                     |
| Blake (1965) [76] |                 | 1.63                        |                |                |                     |
| McKenzie (1980) [60] |                 | 2.75                        |                |                |                     |
| Mewe (2001) [7] |                 | 2.42                        |                |                |                     |
| Behar (2001) [8] |                 | 3.02                        |                |                |                     |
| Xu (2002) [58] |                 | 2.31(18)                    |                |                |                     |
| Ness (2003) [61] |                 | 2.73(57)                    |                |                |                     |
| **Theoretical work** |               |                             |                |                |                     |
| Zhang et al. (1989) [20] | DW | 4.15                        | 12.38          | 16.15          | 3.77               | ./               |
| Bhatia et al. (1992) [77] | RM | 3.74                        | 13.64          | 17.38          | 3.74               | 37              |
| Cornuille et al. (1994) [21] | DW | 4.52                        | 12.63          | 16.07          | 3.44               | 65              |
| Kaastra et al. (1996) [52] | RM | 3.84                        | 13.89          | 17.73          | 3.84               | ./              |
| Safronova et al. (2001) [53] | MBPT | 3.43                       | 10.49          | 14.63          | 4.14               | 36              |
| Dong et al. (2003) [23] | MCDHF | 4.26                       | 10.92          | 15.18          | 4.26               | 20257          |
| Loch et al. (2005) [27] | CI              | 3.91                        | 12.18          | 16.19          | 4.01               | 189            |
| Chen et al. (2007) [24] | Dirac RM       | 3.43                        | 10.63          | 14.81          | 4.18               | ./             |
| Gu (2009) [25] | DW | 4.03                        | 12.33          | 16.23          | 3.90               | ./             |
| Gu (2009) [25] | MBPT | 3.50                        | 10.19          | 14.09          | 3.90               | ./             |
| Jönsson et al. (2014) [26] | CI | 3.56                        | 10.73          | 14.74          | 4.01               | 700000         |
| Santana et al. (2015) [54] | CI | 3.68-3.96                  | 11.58          | 15.71          | 4.13               | 816            |
| Santana et al. (2015) [54] | MBPT | 3.44                        | 10.30          | 14.34          | 4.04               | 7              |
| Oreshkina et al. (2016) [32] | CI | 3.55                        | 10.65          | 14.61          | 3.96               | 100000         |
| Mendoza et al. (2017) [55] | BP-CI | 2.82                        | 9.95           | 15.14          | 5.19               | ./             |
| Wu et al. (2019) [34] | MCDHF | 3.48                        | 10.58          | 14.66          | 4.08               | $3.7 \times 10^6$ |
| Wu et al. (2019) [34] | MCDHF+Breit | 3.56                       | 10.75          | 14.76          | 4.01               | $3.7 \times 10^6$ |
| Kühn et al. (2020) [35] | CI+MBPT (AMBiT [78]) | 3.59(5) | 10.86 | 14.90 | 4.04 | $1 \times 10^6$ |
| Kühn et al. (2020) [35] | CI | 3.49                        | 10.50          | 14.52          | 4.02               | ./             |
| **This work**   | CI              | 3.55(2)                     | 10.71(2)       | 14.74(1)       | 4.03(2)            | $1.2 \times 10^6$ |