



## X-Ray Resonant Photoexcitation: Linewidths and Energies of $K\alpha$ Transitions in Highly Charged Fe Ions

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Photoabsorption by and fluorescence of the  $K\alpha$  transitions in highly charged iron ions are essential mechanisms for x-ray radiation transfer in astrophysical environments. We study photoabsorption due to the main  $K\alpha$  transitions in highly charged iron ions from heliumlike to fluorinelike ( $\text{Fe}^{24+}$  to  $\text{Fe}^{17+}$ ) using monochromatic x rays around 6.6 keV at the PETRA III synchrotron photon source. Natural linewidths were determined with hitherto unattained accuracy. The observed transitions are of particular interest for the understanding of photoexcited plasmas found in x-ray binary stars and active galactic nuclei.

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The spectrum of highly charged iron ions provides rich information on the dynamics of x-ray binary stars [1–11]. Owing to their high transition rate, low intergalactic absorption, and the high relative abundance, the Fe  $K\alpha$  spectral features have been detected in a variety of celestial sources and are of particular interest to the study of high mass x-ray binary stars and active galactic nuclei (AGN) and their accretion disks. The  $K\alpha$  features often provide the last recognizable spectral signature of the accreted material. They are generated by the reflection or absorption of x rays emitted from the vicinity of the compact object or central black hole and, potentially, from the relativistic jets found in AGN and microquasars [12,13]. Hence, not only are properties of the accretion disk encoded in the Fe  $K\alpha$  features, so too are physical properties of the black hole or neutron star. For example, measured Doppler shifts produced by the rotating accretion disk, the relativistic beaming, gravitational bending, and even subtler effects from general relativity, such as those arising from the black hole's spin, all leave imprints on the spectral shape of the Fe  $K\alpha$  transitions [14–19]. A full, quantitative understanding would require not only a consistent modeling of these effects, but also an accurate and complete set of, in the case of photoionized sources, laboratory-tested oscillator strengths and transition energies on which the diagnostics are based.

Experimentally, the Fe  $K\alpha$  transitions have been studied as emission lines from thermal plasmas, in tokamaks [20–22]

and by means of Lawrence Livermore National Laboratory's EBIT-II electron beam ion trap [23,24], using a monoenergetic electron beam. High resolution  $K\alpha$  line spectra of highly charged Ar ions were also obtained with an electron cyclotron resonance ion source [25,26]. In all cases, the excitation of the  $n = 2$  upper level was produced by electron impact excitation, or more complex processes such as inner shell ionization, or dielectronic recombination.

In the present experiment a high-fluence x-ray photon beam is used for excitation, in order to isolate and probe the fundamental structure of highly charged iron ions in the absence of any collisional excitation mechanism. We employ the monoenergetic photon beam produced at the PETRA III synchrotron at DESY to excite  $K\alpha$  transitions in Fe ions, produced and trapped by MPIK's FLASH-EBIT (see Fig. 1). FLASH-EBIT, described in detail by Epp *et al.* [27], was designed for photonic studies of highly charged ions and has been used in several experiments at other synchrotrons and free-electron lasers [28–31]. At its most basic, FLASH-EBIT uses a focused electron beam of several hundred milliamperes to ionize and trap the ions. The electron beam is compressed by means of a magnetic field of 6 T at the trap center to a radius below 50  $\mu\text{m}$ . An electrostatic potential well formed at the center of three drift-tube electrodes provides longitudinal trapping of the ions. In the cylindrical trapping region, an ion cloud of 50 mm length and  $\sim 200 \mu\text{m}$  diameter is stored at an areal density of up to  $10^{11}$  ions/cm<sup>2</sup>. The maximum charge state

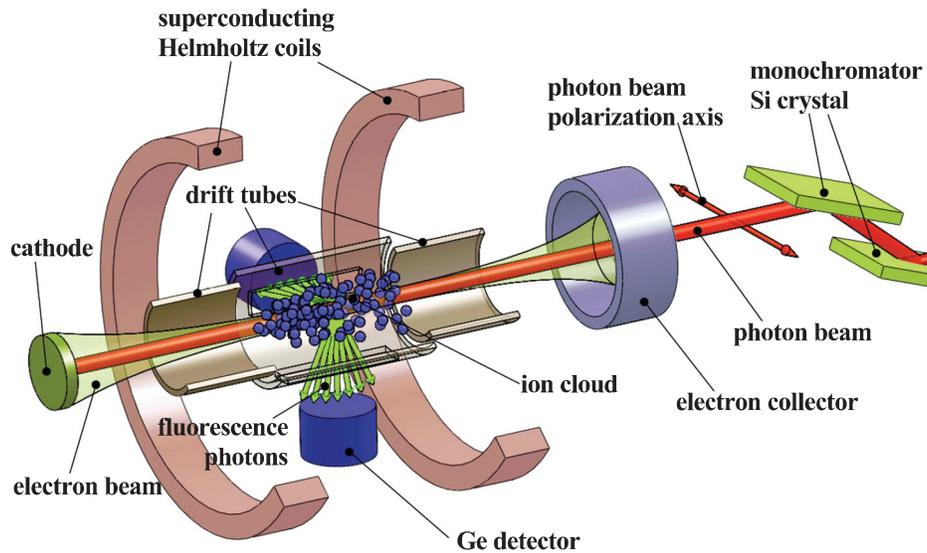


FIG. 1 (color). Scheme of the experimental setup. An electron beam accelerated toward the trap center and compressed by a coaxial 6 T magnetic field produces  $\text{Fe}^{24+}$  ions which are then irradiated by a monochromatic x-ray beam. Resonant excitation leads to fluorescence emission, which is registered by germanium photon detectors.

achievable is predominantly determined by the electron beam accelerating potential, and narrow charge state distributions are routinely achieved. For the experiments presented here, neutral iron atoms are brought into the trapping region by means of a molecular beam of iron pentacarbonyl, which undergoes dissociation and ionization at the crossing point with the electron beam. Once the ions are generated, they are radially trapped by the negative space charge of the beam and axially by the drift tubes. By limiting the electron beam energy to 3.24 keV, the highest charge state produced was  $\text{Fe}^{24+}$ . At this relatively low electron beam energy, direct electron impact excitation of the  $n = 1-2$  transitions in the Fe ions was precluded. Photorecombination-induced background was also absent in the Fe  $K\alpha$  energy region, since the sum of the ionization potential of  $n = 2$  shell vacancies and the kinetic energy of the electron amounts to less than 5 keV. Typical emission spectra with fluorescence photons separated from photorecombination background are shown in Fig. 2.

Trapped ions were resonantly excited using the high-fluence photon beam at the Dynamics Beam line P01 at the PETRA III synchrotron x-ray source. At P01, the ultra-intense x-ray beams are generated using two 5 m long undulators with a period length of 32 mm. Because of the magnetic field orientation of the undulators, a horizontally polarized photon beam is produced. The beam line is dedicated, in general, to inelastic x-ray and nuclear resonant scattering; both methods require high flux. A double-crystal high-heat-load monochromator (HhLM) equipped with two Si crystal pairs is employed. The Si(111) cut crystal yields a resolution of about 1 eV FWHM at 6.6 keV, and with the Si(311) cut 0.2 eV is achieved, albeit with reduced beam intensity. After the HhLM the flux was

up to  $10^{12}$  photons per second on a spot of less than 1 mm diameter and a resolving power of  $E/\Delta E \geq 20000$  was achieved.

The photon beam at P01 was brought into FLASH-EBIT through a beryllium window installed in the electron beam collector chamber. Following our earlier work at advanced light sources, the P01 photon beam is superimposed along

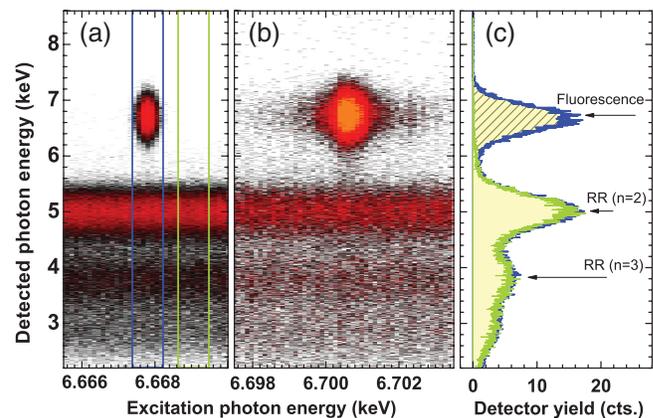


FIG. 2 (color). Typical emission spectra. (a),(b) Detector yield plotted against monochromator energy. Strong signals due to energy dependent fluorescence photons, well separated from energy independent background at lower energies, are visible. On top of the constant background caused by electron impact excitation there is an enhancement at 5 and 3.9 keV. This is due to radiative recombination (RR) by the electron beam into the  $n = 2$  and  $n = 3$  shell. Below 3 keV, strong absorption by the beryllium windows installed in front of the detectors suppresses the signal. (c) The blue curve shows the detector yield in the resonance regime of spectrum (a) and the green curve in a region off resonance of the same width.

the EBIT axis with the trapped ion cloud (Fig. 1). Overlap of the photon beam with the ion cloud was established by use of a retractable scintillator inserted into the trap region and viewed by an image-intensified camera. Both the photon beam and the electron beam fluoresce the scintillator, and maximum overlap is achieved by moving the entire FLASH-EBIT with remote-controlled positioners.

Fluorescent photons are detected using two liquid nitrogen cooled germanium solid-state detectors (1000 mm<sup>2</sup> area each) with an intrinsic resolution of approximately 500 eV FWHM at 6 keV. One detector was aligned parallel and one perpendicular to the x-ray beam polarization axis and both perpendicular to the propagation direction. Beryllium windows mounted radially provided a detection solid angle of  $\Omega \approx 1 \times 10^{-3}$  sr for both detectors. The energy resolution of these detectors allows us to separate the fluorescence signal from the lower photon energy x-ray background generated by electron beam excitation of the trapped ions and by radiative recombination processes.

For these measurements, x-ray fluorescence was measured using the HHLM to scan the x-ray photon beam across the energy band containing the  $K\alpha$  transitions. During the first scans, the energy band between 6530 and 6710 eV was covered using the Si(111) reflection to maximize beam intensity. The observed spectra for both the parallel and the perpendicular detectors are shown in Fig. 3. The spectra include, for heliumlike iron, the  $w$  and  $y$

transitions, for the lithiumlike, the  $q$ ,  $t$ , and  $r$  transitions, and also further resonant transitions in berylliumlike, boronlike, and carbonlike iron ions (Table I). Labels for heliumlike and lithiumlike transitions are used as in Ref. [43]. Because the incident photon beam is polarized, the resulting fluorescence spectra are emitted nonisotropically depending on the transition type. Specifically,  $\Delta J = 1$  transitions have a radiation pattern that peaks at  $90^\circ$  to the polarization axis, while radiation from  $\Delta J = 0$  transitions is isotropic.

To determine transition energies we calibrated the energy scale of the monochromator by measuring the absorption edge spectra of metal foils. The intensity of the photon beam after it passed through each foil was measured using a gas proportional chamber, and represented as a function of the photon beam energy (Bragg angle of the crystal). Digital values of the photon energy selected by the monochromator are provided by a data server in real time. The transmitted photon intensity is normalized to the signal taken before the absorption foil, also using a gas proportional counter. A corrected lattice spacing is used which is more accurate than the control software value by taking into account the crystal's thermal expansion due to the liquid nitrogen cooling [44]. Edges are identified by calculating the photon energy derivative of the transmitted intensity signal. As shown in [45], indirectly determined  $K$ -edge energies for Mn and Fe have 7 and 20 times larger

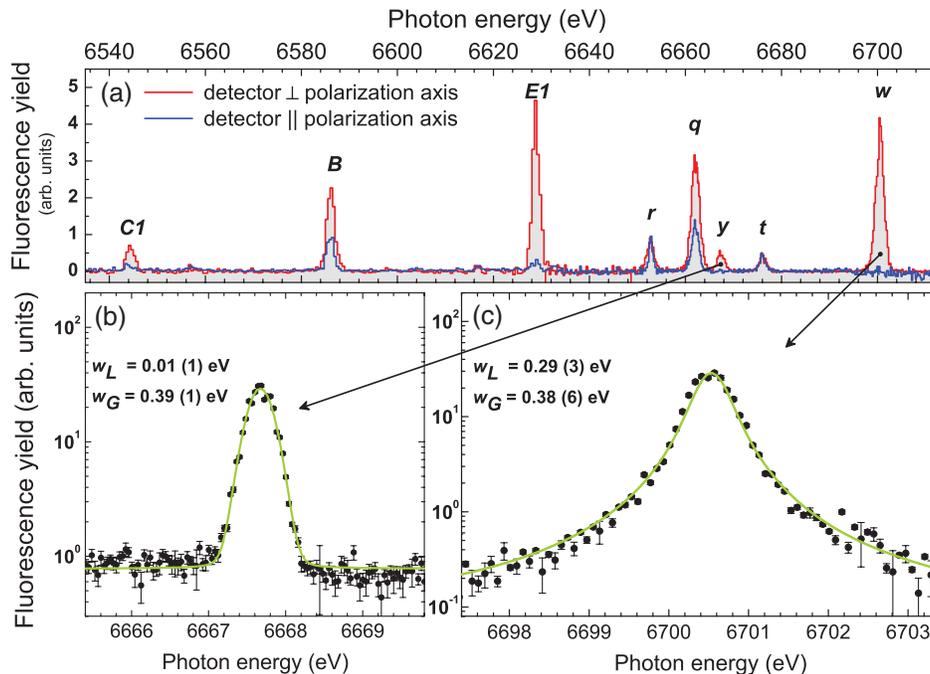


FIG. 3 (color). (a) Overview of the resonance fluorescence spectrum of heliumlike, lithiumlike, berylliumlike, boronlike, and carbonlike Fe ions using two x-ray photon detectors. Alignment of detectors with respect to the polarization of the incoming photon beam: (red curve) perpendicular and (blue curve) parallel. (b) High resolution scan over the intercombination line ( $y$ )  $1s^2\ ^1S_0 \rightarrow 1s\ 2p^3\ P_1$ . (c) High resolution scan over the resonance transition ( $w$ )  $1s^2\ ^1S_0 \rightarrow 1s\ 2p^1\ P_1$ . The fitted Gaussian and the Lorentzian widths  $w_G$  and  $w_L$  from a Voigt profile of the transitions are shown.

TABLE I. X-ray transitions of heliumlike to fluorinelike iron ions resonantly excited from the ground state with synchrotron radiation. X-ray fluorescence was detected as a function of photon energy. Energies are given in units of eV. The calibration is based on the absorption edge technique. The experimental uncertainties are shown as (statistical)(systematic). Relative energies are not affected by the systematic uncertainty which accounts for a shift of the absolute scale. Angle brackets enclose results affected in their accuracy by line blends.

Ion	Line	Initial state	Final state	This experiment	Theory	Theory	Experiment
Fe <sup>24+</sup>	w	1s <sup>2</sup> 1S <sub>0</sub>	1s 2p <sup>1</sup> P <sub>1</sub>	6700.549 (5) (70)	6700.4347 (11) [32] 6700.490 [34]	6700.4 [33] 6700.4 [35]	6700.8 [22] 6700.4 [23] 6700.9 [36]
Fe <sup>24+</sup>	y	1s <sup>2</sup> 1S <sub>0</sub>	1s 2p <sup>3</sup> P <sub>1</sub>	6667.671 (3) (69)	6667.5786 (12) [32] 6667.629 [34]	6667.6 [33] 6667.6 [35]	6667.9 (4) [22] 6667.5 [23] 6667.5 [36]
Fe <sup>23+</sup>	t	1s <sup>2</sup> 2s <sup>2</sup> S <sub>1/2</sub>	1s 2s 2p <sup>2</sup> P <sub>1/2</sub>	6676.202 (3) (69)	6676.129 (47) [37] 6675.8 [38]	6676.4 [33]	⟨6676.8 (7)⟩ [22] 6676.3 [23]
Fe <sup>23+</sup>	q	1s <sup>2</sup> 2s <sup>2</sup> S <sub>1/2</sub>	1s 2s 2p <sup>2</sup> P <sub>3/2</sub>	6662.240 (6) (69)	6662.188 (11) [37] 6661.9 [38]	6661.9 [33]	6662.1 (5) [22] 6662.2 [23]
Fe <sup>23+</sup>	r	1s <sup>2</sup> 2s <sup>2</sup> S <sub>1/2</sub>	1s 2s 2p <sup>2</sup> P <sub>1/2</sub>	6652.826 (3) (69)	6652.776 (25) [37] 6652.6 [38]	6653.5 [33]	⟨6654.2 (7)⟩ [22] 6652.5 [23]
Fe <sup>23+</sup>	u	1s <sup>2</sup> 2s <sup>2</sup> S <sub>1/2</sub>	1s 2s 2p <sup>4</sup> P <sub>3/2</sub>	6616.629 (4) (68)	6616.559 (11) [37]	6616.7 [33]	⟨6617.9 (1.2)⟩ [22] 6616.6 [23]
Fe <sup>22+</sup>	E1	1s <sup>2</sup> 2s <sup>2</sup> 1S <sub>0</sub>	1s 2s <sup>2</sup> 2p <sup>1</sup> P <sub>1</sub>	6628.804 (5) (68)	6631.057 [39] 6627.4 <sup>a</sup> /6628.3 <sup>b</sup>	6628.7 [33] 6627.39 [40]	6628.9 (3) [22] 6628.7 [23]
Fe <sup>22+</sup>	E2	1s <sup>2</sup> 2s <sup>2</sup> 1S <sub>0</sub>	1s 2s <sup>2</sup> 2p <sup>3</sup> P <sub>1</sub>	6597.858 (3) (67)	6596.1 <sup>a</sup> /6597.7 <sup>b</sup> 6596.55 [40]	6595.8 [33]	
Fe <sup>21+</sup>	B	1s <sup>2</sup> 2s <sup>2</sup> 2p <sup>2</sup> P <sub>1/2</sub>	1s 2s <sup>2</sup> 2p <sup>2</sup> P <sub>1/2</sub> 1s 2s <sup>2</sup> 2p <sup>2</sup> D <sub>3/2</sub>	⟨6586.085 (7) (67)⟩ ⟨6586.085 (7) (67)⟩	6586.3 <sup>a</sup> /6585.1 <sup>b</sup> 6587.0 <sup>a</sup> /6585.8 <sup>b</sup> ⟨6587.2⟩ [41]	6586.3 [33] 6586.5 [33]	⟨6585.9 (5)⟩ [22] ⟨6585.7⟩ [23]
Fe <sup>20+</sup>	C1	1s <sup>2</sup> 2s <sup>2</sup> 2p <sup>2</sup> 3P <sub>0</sub>	1s 2s <sup>2</sup> 2p <sup>3</sup> 3D <sub>1</sub>	6544.225 (4) (66)	6544.8 <sup>a</sup> /6544.0 <sup>b</sup>	6543.6 [33]	⟨6544.6 (9)⟩ [22] ⟨6544.4⟩ [23]
Fe <sup>20+</sup>	C2	1s <sup>2</sup> 2s <sup>2</sup> 2p <sup>2</sup> 3P <sub>0</sub>	1s 2s <sup>2</sup> 2p <sup>3</sup> 3S <sub>1</sub>	6556.879 (16) (66)	6557.3 <sup>a</sup> /6556.3 <sup>b</sup>	6555.0 [33]	
Fe <sup>19+</sup>	N1	1s <sup>2</sup> 2s <sup>2</sup> 2p <sup>3</sup> 4S <sub>3/2</sub>	1s 2s <sup>2</sup> 2p <sup>4</sup> 4P <sub>5/2</sub>	6497.067 (5) (65)	6497.5 <sup>a</sup> /6497.2 <sup>b</sup>	6496.6 [33]	⟨6497.7 (1.4)⟩ [22] 6497.3 [23]
Fe <sup>19+</sup>	N2	1s <sup>2</sup> 2s <sup>2</sup> 2p <sup>3</sup> 4S <sub>3/2</sub>	1s 2s <sup>2</sup> 2p <sup>4</sup> 2P <sub>3/2</sub>	6506.845 (7) (65)	6507.3 <sup>a</sup> /6506.9 <sup>b</sup>	6506.0 [33]	⟨6509.6 (1.4)⟩ [22] ⟨6509.1⟩ [23]
Fe <sup>19+</sup>	N3	1s <sup>2</sup> 2s <sup>2</sup> 2p <sup>3</sup> 4S <sub>3/2</sub>	1s 2s <sup>2</sup> 2p <sup>4</sup> 4P <sub>1/2</sub>	6509.133 (14) (65)	6509.6 <sup>a</sup> /6509.1 <sup>b</sup>	6508.1 [33]	⟨6509.6 (1.4)⟩ [22] ⟨6509.1⟩ [23]
Fe <sup>18+</sup>	O1	1s <sup>2</sup> 2s <sup>2</sup> 2p <sup>4</sup> 3P <sub>2</sub>	1s 2s <sup>2</sup> 2p <sup>5</sup> 3P <sub>2</sub>	6466.900 (14) (64)	6467.4 <sup>a</sup> /6466.5 <sup>b</sup> 6466.3 [41]	6564.4 [33]	6467.6 (1.7) [22] ⟨6466.5⟩ [23]
Fe <sup>18+</sup>	O2	1s <sup>2</sup> 2s <sup>2</sup> 2p <sup>4</sup> 3P <sub>2</sub>	1s 2s <sup>2</sup> 2p <sup>5</sup> 3P <sub>1</sub>	6474.318 (33) (64)	6474.9 <sup>a</sup> /6473.9 <sup>b</sup> 6473.7 [41]	6473.0 [33]	⟨6472.7 (2.7)⟩ [22] ⟨6474.7⟩ [23]
Fe <sup>17+</sup>	F	1s <sup>2</sup> 2s <sup>2</sup> 2p <sup>5</sup> 2P <sub>3/2</sub>	1s 2s <sup>2</sup> 2p <sup>6</sup> 2S <sub>1/2</sub>	6435.239 (14) (63)	6435.7 <sup>a</sup> /6434.6 <sup>b</sup>	6434.8 [33]	6436.1 (2.0) [22] 6434.8 [23]

<sup>a</sup>Our theoretical results obtained in the framework of the multiconfiguration Dirac-Fock (MCDF) method [29]

<sup>b</sup>Our theoretical results using the Flexible Atomic Code (FAC) of Gu with the standard configuration-interaction package [42].

uncertainties, respectively, than the direct wavelength measurements of Kraft *et al.* ( $\pm 20$  meV) [46]. Theoretical values given in [45] disagree by 15 eV from the experiment due to unknown solid-state effects. Therefore, we use the *K* edges of Mn, Fe, Co, Ni, and Cu by Kraft *et al.* [46] as references. This results in an angular offset for which one has to correct the Bragg angles provided by the PETRA III data acquisition system since the monochromator has no absolute zero position. In Table I we compare our results with several predictions, which, especially for heliumlike

and lithiumlike iron, agree within the experimental uncertainty with our measurement.

Some of the lines were then investigated at higher resolution in further scans using the monochromator with the Si(311) cut crystal, thereby improving the photon beam resolution by a factor of 5. Additionally, the effect of thermal Doppler line broadening was reduced by cooling the trapped ions evaporatively [47]. We decreased the thermal broadening to be comparable to the natural line-widths of the permitted *1s-2p* transitions while still

TABLE II. Experimentally determined natural linewidths  $w_L$  of several  $1s \rightarrow 2p$  transitions in comparison with predicted values, in units of meV. Uncertainty given as  $1\sigma$ . For line  $B$  we cannot determine a natural linewidth as there is a blend of two transitions. The linewidth of the intercombination lines  $y$  and  $u$  is dominated by Doppler broadening, which also prevents a natural linewidth determination. The estimated uncertainty of MCDF widths is  $\approx 10\%$ .

Line	Experiment	Theory	MCDF		
			Radiative	Auger	Total
$w$	311 (10)	301 [49] 301 [35] 303 [33] 315 [50]	301	0	301
$q$	255 (31)	310 [33]	312	0.081	312
$r$	250 (11)	226 [33]	208	32	241
$t$	131 (29)	167 [33]	112	52	163
$E1$	437 (12)	382 [33]	292	95	387
$E2$	178 (34)	149 [33]	31	136	167
$C1$	524 (12)	499 [33]	218	334	552
$C2$	385 (207)	611 [33]	412	135	547
$N1$	565 (45)	498 [33]	146	400	546
$N2$	594 (50)	504 [33]	164	338	502
$N3$	570 (109)	505 [33]	155	360	515
$O1$	859 (229)	756 [33]	264	529	793
$O2$	772 (228)	785 [33]	304	518	822
$F$	998 (203)	989 [33]	351	651	1020

producing sufficient fluorescence signal. High resolution scans of  $w$  and  $y$  are shown in the bottom panels of Fig. 3. Natural linewidths were determined by fitting the line shapes with a Voigt profile, as done by Beiersdorfer *et al.* [48] for  $\text{Cs}^{45+}$  under electron impact excitation. A weighted average of the Lorentzian widths  $w_L$  in all energy scanned spectra is taken. The intercombination transition  $^3P_1 \rightarrow ^1S_0$  in heliumlike Fe forming line  $y$  has a negligibly small natural linewidth in comparison to the Doppler width of this line. We compare the Doppler widths (Gaussian widths of the Voigt profiles) of the heliumlike line  $w$  with the linewidths of  $y$  for spectra taken at the same EBIT conditions and find perfect agreement within the error bars, which supports our results for the natural linewidths. Our natural linewidth measurements are summarized in Table II, which also displays our theoretical results obtained in the framework of the multiconfiguration Dirac-Fock (MCDF) method [29]. The good agreement with theory extends even to the more complex ions, for which one might have expected larger deviations.

The strong  $K\alpha$  transitions of the highly charged iron ions are essential for astrophysical plasma diagnostics. With the present measurements, their resonant photoabsorption has been directly measured, and state-of-the-art predictions for both their transition probabilities and energies have been benchmarked against, and confirmed by,

them with satisfactory results. As for the transition energies, our systematic uncertainty enlarges the total final error bar to a typical level of 70 meV while the energy differences show 10 times smaller error bars. These results will be particularly useful when interpreting astrophysical x-ray spectra, especially from active galactic nuclei and high mass x-ray binary stars, where absorption and reflection of x rays from an accretion disk is directly related to the physical properties of potentially every physical property of the system. The data will be especially useful for benchmarking theoretical calculations of atomic structure and spectral models used to interpret spectra from celestial sources, such as XSTAR [51] and CLOUDY, and are timely given the launch of the x-ray calorimeter spectrometer [52] on ASTRO-H [53] in 2015.

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