

Magnetic-Field Induced Enhancement in the Fluorescence Yield Spectrum of Doubly Excited States in Helium

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An influence of static magnetic fields on the fluorescence yield spectrum of He in the vicinity of the $N = 2$ thresholds has been observed. The experimental results are in excellent agreement with predictions based on multichannel quantum defect theory, and it is demonstrated that the Rydberg electron ℓ mixing due to the diamagnetic interaction is essential for the description of the observed fluorescence yield intensity enhancement.

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Doubly excited states of helium are fundamental for the understanding of electron correlation and have been extensively studied ever since their discovery [1]. The photoabsorption spectrum measured in the pioneering synchrotron radiation (SR) work by Madden and Codling [2] has a complexity which is further explored with every new experimental improvement [3]. Much can be learned also in experiments where the yield of secondary photons [4–7] or metastable atoms [8] is measured as the energy of the primary photons is varied over the double-excitation resonances. It has been demonstrated that the fluorescence yield (FY) spectrum close to the $N = 2$ thresholds is determined by a subtle balance between the radiative and autoionizing channels [6]. This balance makes FY spectra very sensitive to external fields, so that a strong influence on the intensity can be observed also with electric fields much weaker [9,10] than the fields required for direct measurements of Stark shifts [11].

Influence on the double excitations of magnetic fields has been discussed in connection with dielectronic recombination (DR) [12,13], and then mostly when magnetic and electric fields are combined. The intrinsic influence of magnetic fields on the doubly excited states of helium has until now remained experimentally unexplored, most likely because it has been believed that the moderate fields feasible in laboratory experiments would not have a noticeable effect on the spectra. Early theoretical results indicated that pure magnetic fields below 5 T should not have any influence on DR cross sections [14]. We show here that the fields generated by a conventional electromagnet (up to 1 T) strongly influence the FY spectrum. The B -field influence on the FY spectrum is well described within multichannel quantum defect theory (MQDT), demonstrating that the principal B -field dependence is due to

the diamagnetic interaction. The diamagnetic term in the Hamiltonian is proportional to B^2 and gives an effect at radically lower interaction strengths (~ 25 times) than has earlier been expected. Through this interaction the B field mixes dipole allowed states with states that are forbidden in the field-free case, much in the same way as electric fields. As these states have larger fluorescence branching ratio (FBR) than the dipole allowed states, this leads to the observed FY intensity enhancement.

The experiment used linearly polarized SR with an energy resolution of around 1.6 meV at the gas-phase beam line at ELETTRA [15]. Helium gas was let in to a differentially pumped gas cell at a pressure of $\sim 10^{-3}$ torr in a chamber with a pressure of 10^{-6} torr. The cell was mounted on the poles of an electromagnetic yoke, in the gap of which the interaction region was situated (Fig. 1). The direction of the magnetic field was parallel to polarization direction of the incoming photon beam. By adjusting the current in coils mounted outside the vacuum the B field in the interaction region could be varied up to 1 T. The dependence of the magnetic field on the electrical current through the coils was determined *ex situ* using a Hall probe, and simulations indicate that the field was homogeneous within a few percent.

Fluorescence was measured in a direction perpendicular to the plane of polarization of the incoming radiation with a detector based on multichannel plates (Fig. 1). The detector was separated from the gas by a filter, which ensured that charged particles and metastable atoms could not reach the detector. The filter material (1500 Å aluminum, 300 Å carbon) was chosen to give high transmission around 40 eV ($\sim 20\%$), corresponding to the radiative decay of the doubly excited states, while blocking secondary emission at energies below 25 eV (transmission $< 1\%$).

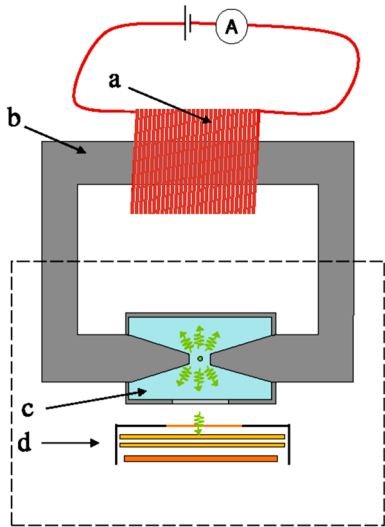


FIG. 1 (color online). Schematic view of the experimental setup. (a) Coils; (b) magnetic yoke; (c) gas cell; (d) multi-channel plate detector. The SR enters perpendicularly to the plane of the figure in the center of the gas cell, with its polarization in the direction of the field defined by the magnetic poles. The dashed line represents the vacuum envelope.

At lower energies in Fig. 2, the $B = 0$ spectrum is dominated by the stronger n^+ series (beginning with 13^+ at around 65.32 eV) interspersed with the unresolved $n^0/(n+1)^-$ series (beginning with $13^0/14^-$ just above 13^+), where n^+ , n^- , n^0 is a short notation for the states represented [16] by the LS allowed $(2snp + 2pns)^1P_1^o$, $(2snp - 2pns)^1P_1^o$, and $2pnd^1P_1^o$ states, respectively. As we have pointed out earlier [5,6] LS coupling cannot account for the intensity close to the $N = 2$ thresholds, where mixing with spin-forbidden states becomes substantial. As these states have larger FBR than the LS allowed states this results in an intensity enhancement in the FY spectrum in the region closer to threshold where the individual peaks remain experimentally unresolved. Here the

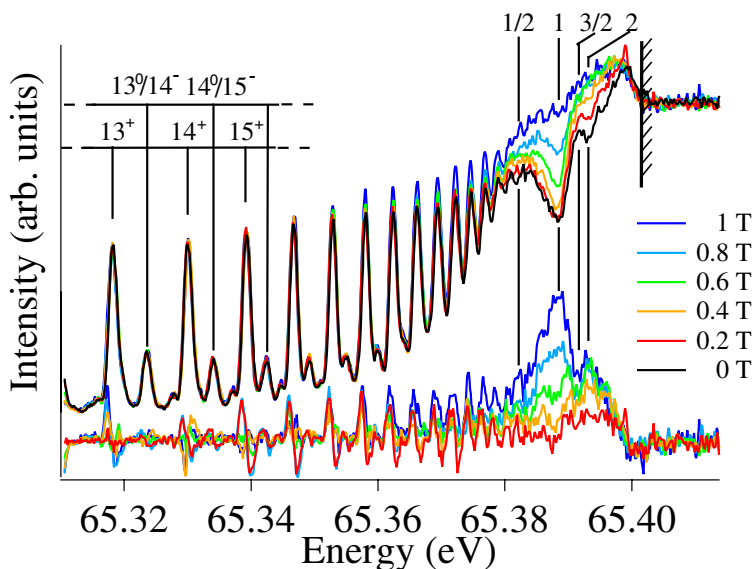


FIG. 2 (color online). Fluorescence yield spectra of helium in magnetic fields varying in steps of 0.2 T up to 1 T. Some n^+ , n^- , and n^0 states are labeled for orientation at lower energies, and vertical lines indicate $\Delta\nu = \frac{1}{2}, 1, \frac{3}{2}, 2$ for the field-free spectrum closer to threshold. The spectra are normalized by assuming that the FY intensity above the $N = 2$ thresholds is independent of the applied field. Increasing B monotonically increases intensity in the 65.38–35.39 eV region, and in general induces increasing deviations from the free field spectrum. To highlight the variation, $FY(B) - FY(0)$ difference spectra, where the $B = 0$ spectrum has been subtracted from the spectra at each B field, are shown in the lower part of the figure.

intensity variations are reproduced by calculations in the JK coupling scheme [6], demonstrating a crucial dependence on the effective quantum numbers, ν_j , associated with Rydberg series converging to the different spin-orbit split $2p_{1/2}$ and $2p_{3/2}$ thresholds. At energies where the difference between the quantum numbers, $\Delta\nu$, is an integer the FY intensity is as small as predicted by LS coupling, whereas when $\Delta\nu$ is a half integer maximum departure from LS coupling and hence maximum enhancement in FY intensity is attained [6]. In Fig. 2 this is reflected as local intensity minima at $\Delta\nu = 1, 2$, and maxima at $\Delta\nu = \frac{1}{2}, \frac{3}{2}$ in the $B = 0$ spectrum.

The FY intensity increases with applied magnetic field, especially in the 65.38–65.39 eV region (Fig. 2). For $B = 1$ T a significant enhancement is seen already at 65.35 eV (around 18^+) and it becomes most pronounced around 65.389 eV, while the spectrum is unaffected by the B field a few meV below the thresholds at 65.40 eV. To further visualize the magnetic-field dependence we subtract the $B = 0$ spectrum and show also the residuals in Fig. 2. Note that there are significant effects on the spectrum at very small fields, readily measurable at $B = 0.2$ T.

The energy stability of the monochromator puts a limit to the accuracy at steep intensity variations, and the asymmetric line profiles at lower energies in the difference spectra may be due to shifts in the monochromator energy over time rather than B -field induced shifts.

Obviously, it is just at energies where the LS result is retrieved in the field-free spectrum that the enhancement induced by the B field is particularly strong. The difference spectra have local *maxima* when $\Delta\nu$ is an integer. As the corresponding states have relatively small FBR in the field-free spectrum any state mixing is likely to increase the FY intensity. This is also in line with the observation that the spectra are unaffected the last few meV below the thresholds: at such high Rydberg states $FBR \sim 1$ for all states and consequently no further FY intensity enhancement can be induced via this mechanism.

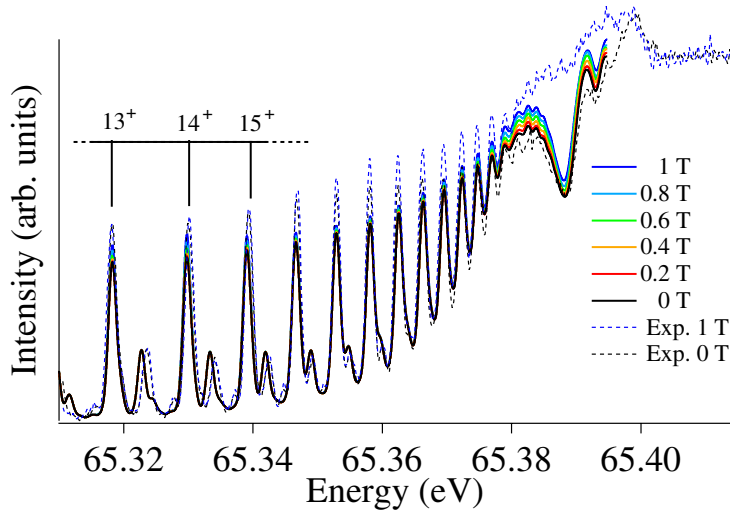


FIG. 3 (color online). Theoretical FY spectra of helium at magnetic fields varying in steps of 0.2 T up to 1 T, only taking the paramagnetic shifts of the $N = 2$ thresholds into account. The theoretical results are convoluted with a Voigt function, determined by a fit to the resolved peaks at lower energies in the experimental spectra. For comparison we show also the experimental spectra for $B = 0$ and 1 T. The theoretical and experimental spectra are normalized so that the $B = 0$ spectra coincide in the intensity dip at $\Delta\nu = 1$. Increasing B monotonically increases the theoretical FY intensity.

To interpret the magnetic-field effects on the spectra in detail we use MQDT. The calculation proceeds in a manner somewhat similar to the calculation of FY in a static electric field [9]. We use the 0-field K matrices and dipole matrix elements as input into a procedure that incorporates the full effect of the magnetic field. The 0-field channels with different total angular momenta J are coupled together by the magnetic field. There are two competing effects. (1) A shift in the channel thresholds due to a paramagnetic shift in the channel energies. (2) A mixing of the ℓ quantum numbers of the Rydberg electron due to the diamagnetic, $B^2(x^2 + y^2)$, term in the Hamiltonian.

To account for these shifts and mixings, we need to uncouple the angular momenta. The 0-field channels are specified by the angular momentum of the core j_c , the angular momentum of the outer electron j_o , and the total angular momentum J ; at this stage the channels are specified as $|j_c, j_o, JM_J\rangle$. First, we use Clebsch-Gordon coefficients to uncouple the angular momentum of the core and outer electron; now the channels are specified as $|j_c, m_c, j_o, m_o\rangle$ where $m_c + m_o = M_J$. Then we use Clebsch-Gordon coefficients to uncouple the spin of the outer electron from the orbital angular momentum of the outer electron; now the channels are specified as $|j_c, m_c, s_o, \mu_o, \ell, m_\ell\rangle$ where $m_c + \mu_o + m_\ell = M_J$. The core energy for each channel shifts by an amount $(g_c m_c + 2\mu_o + m_\ell)B/2$ where $g_c = 2$ for the $2s_{1/2}$ state, $g_c = 4/3$ for the $2p_{3/2}$ state, and $g_c = 2/3$ for the $2p_{1/2}$ state.

At $B = 1$ T these shifts are on the same order of magnitude as the $2p_{3/2}$ - $2p_{1/2}$ splitting ($727 \mu\text{eV}$) in the $B = 0$ case. The latter splitting determines the $\Delta\nu$ parameter which is crucial for the description of the $B = 0$ spectrum close to the $N = 2$ thresholds. With such big B field induced shifts the relative spacing of Rydberg levels changes substantially at these energies, and one would intuitively expect that $\Delta\nu$ loses its meaning and hence significant spectral changes. In Fig. 3 we show calculated FY spectra where we only include the shift in thresholds from the magnetic field. Indeed a FY enhancement is

predicted in the investigated energy region. The effect on the FY cross section is, however, much smaller than observed, and obviously much larger B fields are required before this effect becomes important. Thus, we conclude that the paramagnetic effect alone cannot account for our experimental results.

The diamagnetic term is proportional to $B^2(x^2 + y^2)$ and it therefore increasingly mixes ℓ as the principal quantum number increases, much in the same way as the ℓ mixing due to a static electric field [9]. Using 0th order perturbation theory for hydrogen, the states within an n manifold is predicted to have an energy spread up to $(5/16)n^4[B/(2.35 \times 10^5 T)]^2$ [17]. This spread is of similar size as the spread induced by the threshold displacements. In our calculations of FY in a static electric field [9] we accounted for the ℓ coupling for each m_ℓ from the field by performing a further transformation from spherical to parabolic coordinates for the Rydberg electron. Unfortunately, there is no transformation available for strong magnetic fields. To incorporate the ℓ coupling, we used a method similar in spirit to that in Ref. [18]. We diagonalized the diamagnetic Hamiltonian in a basis set to obtain the Green's function near the origin. Using a method comparable to the one used in Ref. [19] we were able to close the channels and obtain the FY spectrum in the field from this Green's function. Because the diamagnetic term in the Hamiltonian only couples ℓ to $\ell \pm 2$, there are only $24 M_J = 0$ channels of the type $1s\epsilon p$, $2snp$, $2pns$, and $2pnd$ with nonzero K matrix elements that are used in the calculation.

In Fig. 4 we demonstrate that the agreement between the experimental results and the theoretical predictions is excellent. Even details emphasized in the difference spectra are accounted for. We can thus conclude that the theoretical treatment as outlined above captures the essential mechanism for the observed B -field induced FY enhancement. This result also implies that the diamagnetic interaction, rather than the paramagnetic, is crucial for the spectral B field dependence for $B \leq 1$ T.

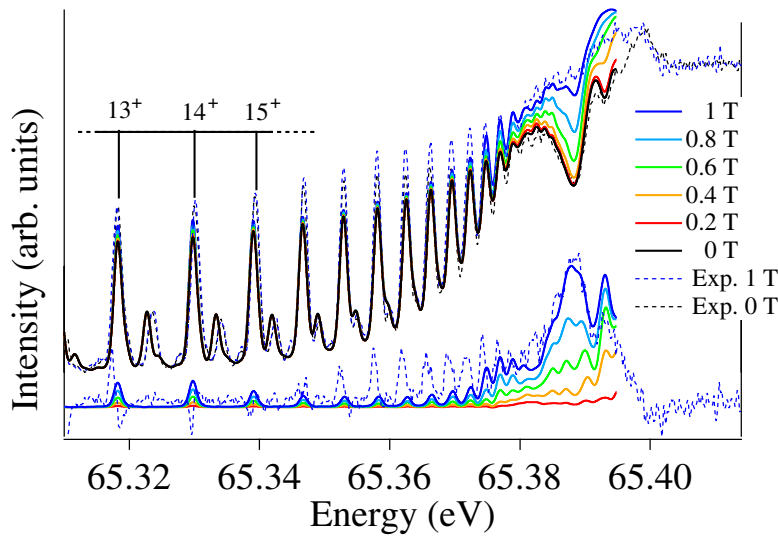


FIG. 4 (color online). Theoretical FY spectra of helium at magnetic fields varying in steps of 0.2 T up to 1 T, using the full theory including diamagnetic ℓ mixing and paramagnetic shifts of the $N = 2$ thresholds. For comparison we show also the experimental spectra for $B = 0$ and 1 T. The theoretical and experimental spectra are normalized so that the $B = 0$ spectra coincide in the intensity dip at $\Delta\nu = 1$. Increasing B monotonically increases the theoretical FY intensity. The lower part of the figure shows calculated $FY(B) - FY(0)$ difference spectra, and for comparison the experimental $FY(B) - FY(0)$ difference spectrum for $B = 1$ T is shown as a dashed line.

In general the biggest magnetic-field influence on the FY spectrum is due to mixing with states that have larger FBR than the states reached in the field-free case, in analogy to the electric-field effects [9]. However, whereas the eigenenergies fan symmetrically out from each n manifold with increasing electric field, the energies for the diamagnetic effect fan only upward. This implies that low m_ℓ states do not mix in due to the B field until the region where n mixing becomes important is reached.

These results have implications for the understanding of processes where the FBR of the doubly excited states is important, e.g., it implies an enhancement of the DR cross sections. This may be difficult to observe directly in DR experiments at ion storage rings, but it could be important when considering DR processes in plasmas, both of astrophysical relevance, and of relevance for attaining terrestrial fusion. In both cases the magnetic fields are often much higher than those investigated here. Note also that the magnetic-field induced increase of FBR implies a reduction in the competing autoionization branching ratio, and thus a magnetic-field induced reduction of intensity in electron yield spectra. This should be readily observable with modern SR techniques. In general, measurements of x-ray fluorescence in high magnetic fields is an area of growing interest at SR facilities.

We expect similar behavior for He-like highly charged ions, although the magnitude of the required B field remains to be clarified. The important parameters are the spin-orbit splitting which is determined by $(Z/2)^4 E_{\text{so}}$, where E_{so} is the spin-orbit splitting for He^+ . The position of the intensity dip in the field-free FY spectrum can be estimated by $(Z-1)^2/\nu^3 = (Z/2)^4 E_{\text{so}}$, and hence it is in general predicted to be at $\nu = [(Z-1)^2(2/Z^4)]^{1/3} \nu_{\text{He}}$. Because the size of the diamagnetic interaction goes like $B^2 \nu^4/Z^2$, this suggests that larger B fields are needed. However, the size of the quantum defects also goes down with Z , which might counteract this effect.

In conclusion, we have presented the dependence on a static external B field of the FY spectrum of the doubly

excited states of He close to the $N = 2$ thresholds. Effects induced by the B field are found to be much larger than earlier expected. Mixing of states with larger FBR than the states reached in the field-free case leads to a dramatic intensity enhancement in the FY spectrum. The experimental results are excellently described by predictions based on MQDT, showing that the diamagnetic interaction is the main reason for the observed intensity enhancement.

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