Fine Structure of the $1s4f$ Configuration of Helium

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(Received 18 May 1987)

By use of an rf electric resonance technique, the fine structure of the $1s4f$ multiplet of He I and its splitting in a static electric field have been investigated. Our results for the fine-structure intervals ν_{1J} between the ${}^1F_3$ level and the ${}^3F_J$ levels are $\nu_{12}=232.8(2.0)$ MHz, $\nu_{14}=490.8(2.0)$ MHz, and $\nu_{13}=704.3(1.0)$ MHz. Furthermore, a singlet-triplet mixing ratio $\Omega=0.757(3)$ and the exchange interaction $2K=158(3)$ MHz have been derived from the measurements.

PACS numbers: 35.10.Fk, 32.30.Bv

The fine-structure (fs) splittings of most singly excited He ($1snl$) levels have been measured[1a] with high precision. The fs of configurations with $l \leq 2$ has been investigated by magnetic resonance,[1b] level crossing,[2] quantum beat,[3] and anticrossing techniques,[4] as well as by Doppler-free laser spectroscopy.[5] Multiplets with $l \geq 2$ have been analyzed by microwave-optical resonance measurements.[6,7] So far, none of these techniques has been suitable for measuring the fs of the $1s4f$ configuration. Nevertheless, this configuration has attracted much interest due to the fact that it is the energetically lowest level of He I with a significant mixing of singlet and triplet states. Several theoretical investigations[8-11] of its electric and magnetic fs have been prompted by the observation of apparently spin-forbidden transitions in thermal He($1s^2$)-He*($1snp\ {}^1P$) collisions, after St. John and Fowler[12] proposed that these transitions are made possible by the singlet-triplet mixing in $1snl$ configurations with $l \geq 3$. Further interest in the $1s4f$ configuration has been stimulated by the observation that He triplet states are excited in p-He collisions[13] and He^{++}-He collisions.[14] In a series of experiments[15] it has been demonstrated that these excitation processes proceed predominantly via the $1s4f$ configuration.

In spite of the fundamental importance of the singlet-triplet mixing in $l \geq 3$ levels, the mixing ratios are not well known. Experimentally, they can be determined accurately by studying the splitting of fs levels in external electric or magnetic fields. However, to our knowledge such measurements have not been performed for these levels. Only for the $1s4f$ levels have some crude cascade level-crossing measurements been published.[16] Theoretically, the evaluation of the exchange interaction presents the main obstacle to a reliable evaluation of the mixing ratio. The published calculated values[8-11] are not in agreement (see Table I).

In this Letter, we report the first measurement of the fs intervals of the $1s4f$ multiplet in He I and of the splitting of these levels in an external electric field. An accurate value for the mixing coefficient has been determined from these measurements. The experiments are based on an electric resonance technique.[17] For excitation, we made use of 20-keV deuteron impact to excite singlet states selectively according to Wigner's spin-selection rule.[18] To detect the resonance signals, we observed the visible spectral lines $\lambda(1s3d\ {}^1D-1s2p\ {}^1P)=668$ nm or $\lambda(1s3d\ {}^3D-1s2p\ {}^3P)=588$ nm emitted in the cascade decays $1s4f$-$1s3d$-$1s2p$. Radio-frequency transitions within the $1s4f$ multiplet were induced by an rf electric field as proposed recently.[17] As a result of the large polarizabilities α of He I states with $l \geq 2$, it is more favorable to induce two-quantum electric-dipole ($2E1$) transitions than magnetic-dipole transitions. If in addition to the rf electric field E_1 a static electric field E_0 is also applied, one-quantum electric-dipole ($1E1$) transitions also can be induced as a result of the mixing of states with opposite parity. The Rabi frequencies are of the order of $\omega_1 \simeq \alpha E_1^2/4$ for $2E1$ and $\omega_1 \simeq \alpha E_1 E_0$ for $1E1$ transitions, where α is typically of the order of $\alpha \simeq 100$ Hz/(V/cm)^2 for the $1s4f$ states. These techniques made it possible to induce rf transitions within the $1s4f$ multiplet in spite of its rather short radiative lifetime $\tau(1s4f)=74$ ns.[16]

The experimental setup used was essentially the same as in Ref. 17. The most important part was the resonance cavity shown in Fig. 1. The (not mass-selected) ion beam was extracted from a Penning ion source with a D_2 gas inlet and passed through the evacuated lower part of the cavity–He-gas target. The upper electric-field plate P_1 was grounded through an inductor L_1, and the lower plate P_2 (insulated from the bottom plate of the

TABLE I. Comparison of the measured singlet-triplet mixing ratio Ω and the corresponding exchange interaction $2K$ (in megahertz) with theoretical predictions.

	Ω	$2K$
This work	0.757(3)	158(3)
Ref. 10		137.5
Ref. 8	0.4335[a]	601.6[a]
	0.6151[b]	337.4[b]
Ref. 9	0.593	

[a] With polarization correction.
[b] Without polarization correction.

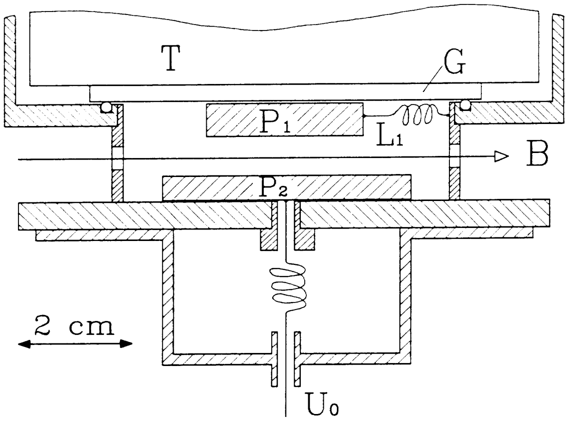

FIG. 1. Lower part of the resonance cavity where the ion beam B passes through the He-gas target. The static electric field E_0 is produced by applying a voltage U_0 between the stainless-steel plates P_1 and P_2. The rf electric field E_1 is produced by exciting the tank circuit tuned to various resonant frequencies $100\ \mathrm{MHz} \lesssim \nu \lesssim 1000$ MHz by placing the corresponding tuning inserts T into the upper part of the cavity, which is at atmospheric pressure and separated from the lower part by a glass plate G.

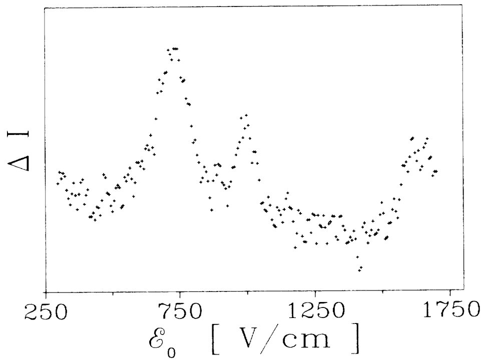

FIG. 2. Signals of resonance transitions induced by an rf field E_1 at $\nu = 397.4$ MHz scanned by varying the static-electric-field strength E_0. The signal is the intensity change ΔI of the $\lambda = 668$-nm line caused by the rf transitions and measured by lock-in detection.

cavity by a 0.1 mm Teflon foil) was connected to a dc voltage supply by a low-pass filter to produce electric fields $0 < E_0 \lesssim 1700$ V/cm. Impact radiation emitted perpendicular to the plane of the drawing was observed through an aperture (diameter 1 cm) in the wall of the cavity. The upper part of the cavity was a tank circuit tuned to various resonant frequencies $100\ \mathrm{MHz} \lesssim \nu \lesssim 1000$ MHz by replaceable inserts. The resonator was capacitively coupled to an rf generator. Both the static field E_0 and the rf field E_1 were measured using level-crossing signals[19] from the $\lambda(1s4d\,^1D - 1s2p\,^1P) = 492$ nm and the $\lambda(1s5d\,^1D - 1s2p\,^1P) = 439$ nm lines. Based on the well-known tensor polarizabilities of the $1snd\,^1D$ levels,[17] the ratio $U_0/E_0 = 0.705$ cm was determined to an accuracy of about 1%, and E_1 was typically $E_1 \approx 50$ V/cm for $P = 2$ W input power.

Resonance signals (Fig. 2) were detected by keeping the frequency of the rf field fixed and varying the applied voltage U_0 typically from 20 to 1200 V. The electric-field strengths for all resonance signals observed at frequencies in the range $150\ \mathrm{MHz} \lesssim \nu \lesssim 1000$ MHz are plotted in Fig. 3. We interpret the resonance signals using the splitting scheme of the $1s4f$ multiplet in a static electric field (Fig. 4). Since E_1 and E_0 are parallel to one another, only $\Delta m = 0$ transitions can be induced. The strongest signals are expected for transitions from substates with predominantly singlet character to those with triplet character. At low electric-field strengths, the following transition frequencies have been identified (see Fig. 4): $\nu_{13}(1)$, $\nu_{14}(1)$, $\nu_{14}(3)$, $\nu_{12}(1)$, and $\nu_{12}(1)/2$, $\nu_{14}(3)/2$. Here $\nu_{1J}(|m|)$ refers to a transition from the singlet sublevel $4\,^1F_3(|m|)$ to the triplet sublevel $4\,^1F_J(|m|)$ with Stark quantum numbers $0 \le |m| \le J$. The first four frequencies correspond to $1E1$ transitions, and the last two correspond to $2E1$ transitions. At high electric-field strengths, where the angular momenta L and S are almost decoupled, resonances were observed for the following transition frequencies: $\nu_{13}(1)$, $\nu_{14}(1)$,

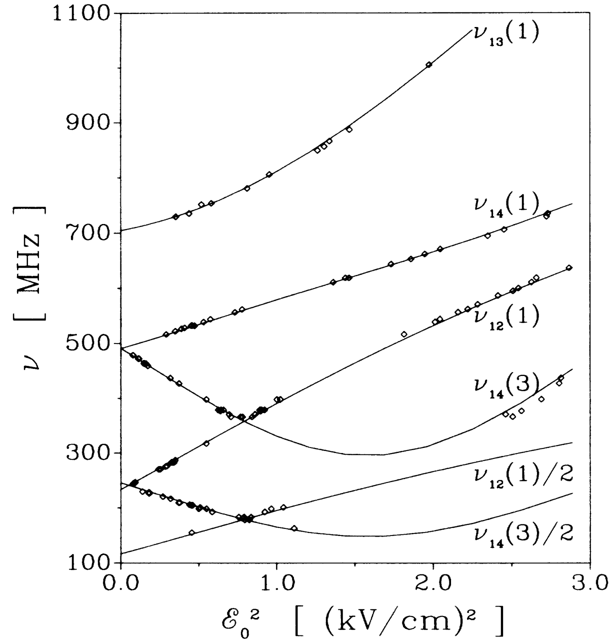

FIG. 3. Electric-field strengths E_0 of resonance signals detected with rf fields at frequencies $150\ \mathrm{MHz} \lesssim \nu \lesssim 1000$ MHz. The experimental results are compared with calculated resonance frequencies $\nu_{1J}(|m|)$ (solid lines) of transitions from a $^1F_3$ sublevel to a ^3F_J sublevel with $0 \le |m| \le J$.

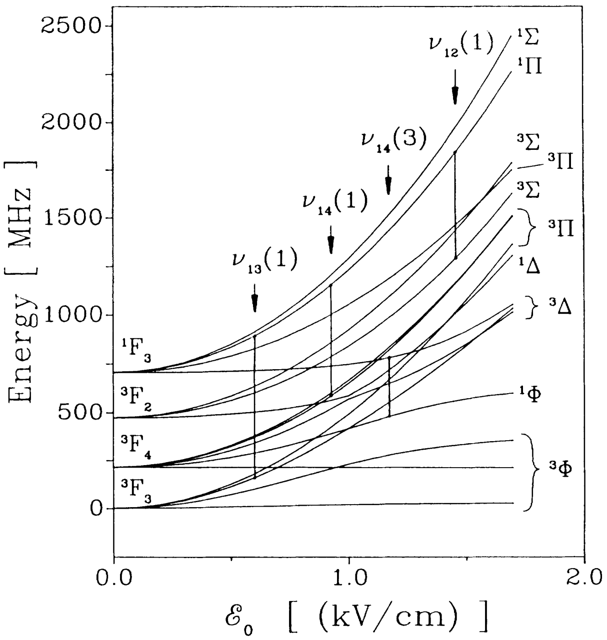

FIG. 4. Splitting scheme of the $1s4f$ multiplet in a static electric field E_0. Detected transitions $\nu_{1J}(|m|)$ are indicated. The asymptotic quantum numbers of the high-field multiplets $^1\Lambda$ and $^3\Lambda$ with $0 \le \Lambda = |m_L| \le 3$ are given.

$\nu_{12}(1)$, and $\nu_{14}(3)$. According to the splitting scheme (Fig. 4), these resonances correspond to transitions from $^1\Lambda$ to $^3\Lambda$ substates, where Λ is the asymptotic high-field quantum number $|m_L|$.

The zero-field fs splittings can be evaluated by extrapolating the observed low-field resonance frequencies for $E_0 \to 0$, yielding $\nu_{13} \approx 705$ MHz, $\nu_{14} \approx 490$ MHz, and $\nu_{12} \approx 232$ MHz. Furthermore, the mixing ratio Ω of singlet and triplet states at zero electric field can be deduced from the electric-field dependence of the resonance frequencies. Defining $\Omega = \tan\vartheta$ as usual,[16] using the mixing angle ϑ of the eigenstates $|1s4f\,^1F_3,m\rangle$ and $|1s4f\,^3F_3,m\rangle$ with respect to the Russell-Saunders basis states, we obtained $\Omega \approx 0.75$ from the observed low-field frequency shifts.

A detailed analysis of the entire set of resonance signals (Fig. 3) was performed within the framework of the theoretical approach of Cok and Lundeen.[11] Using the magnetic coupling constants $h_{so} = -32.509$ MHz, $h_{off} = 97.841$ MHz (spin-orbit coupling) and $h_{ss} = 32.598$ MHz (spin-spin coupling) calculated by Cok and Lundeen[11] to a precision of about 0.1%, and using polarizabilities of the Russell-Saunders basis states calculated in the hydrogenic approximation to an accuracy of better than 1%,[16,20] there remains only the mixing ratio Ω (or alternatively the exchange interaction $2K$) as a free parameter for fitting the calculated level splittings to all the measured resonance frequencies. Within the limits of experimental errors, a satisfactory fit was obtained for

$$\Omega = 0.757 \pm 0.003.$$

TABLE II. Measured and calculated fs intervals $\nu_{1J} = |E(^1F_3) - E(^3F_J)|/h$ of the $1s4f$ multiplet (in megahertz) together with corresponding singlet-triplet mixing ratios Ω.

	Ω	ν_{12}	ν_{14}	ν_{13}
Ref. 6		215.7	481.7	663.3
Ref. 11	0.75	237.0	495.0	706.1
	0.76	231.1	489.1	703.5
Present work	0.757(3)	232.8(2.0)	490.8(2.0)	704.3(1.0)

In Table II, our final results for the transition frequencies of the $1s4f$ multiplet deduced from this fit are compared with the "global fit" values[6] extrapolated from microwave measurements on $1snf$ multiplets with $n \ge 7$.[21] The fs intervals calculated with $\Omega = 0.75$ and 0.76 using the theory of Cok and Lundeen[11] demonstrate how sensitively these intervals depend on the mixing ratio.

Finally, the exchange interaction

$$2K = \nu_{13}\cos(2\vartheta) - h_{so} - 2h_{ss} = (158 \pm 3)\ \text{MHz}$$

was evaluated from the deduced Ω. This experimental value can be compared with the previously mentioned theoretical predictions (Table I). Using a simple hydrogenic approximation,[8,9] one obtains an exchange integral roughly twice as large as the experimental value (row 4). Taking into account a polarization correction[8] leads to an even larger exchange interaction (row 3). By including a second-order exchange interaction, Chang and Poe[10] have obtained a value (row 2) of the same order of magnitude as our result, although the agreement is still not satisfactory.

In conclusion, we emphasize that our results have confirmed the basic idea of the theoretical approach of Cok and Lundeen[11]: that the magnetic fs interaction of $1snl$ states of He I with $n \ge 3$ can essentially be evaluated within Heisenberg's hydrogenic approximation.[22] However, the exchange interaction is still poorly understood. The best theoretical value[10] deviates from the experimental one by 13% in the case of $1s4f$. From zero-field fine structure intervals[6] one derives a discrepancy of 9%–11% for the higher $1snf$ states.[11]

The rf electric resonance technique has proved to be a valuable tool for spectroscopic investigations of short-lived excited helium states. Combined with a thorough analysis of the amplitudes of the resonance signals, it may also become helpful for investigations of ion-impact excitation of helium atoms, in particular for the determination of excitation matrices of triplet states and for the analysis of cascade effects, where level-crossing techniques[23] are less suitable.

We would like to thank R. Müller for his computational assistance. This work has been supported by the

Deutsche Forschungsgemeinschaft under Contract No. Op20/6-1.

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[21] The predictions of the "global fit" of Farley, MacAdam, and Wing (Ref. 6) deviate from the measured fine structure intervals of the $1s6f$ and $1s5f$ configuration of Farley *et al.* (Ref. 6) by up to 2 and 5 MHz, respectively. The discrepancies are much larger (up to 41 MHz) for the $1s4f$ configuration. These deviations can be reduced by changing the fitting parameters of the "global fit," but might also be due to the inadequacy of the fitting formula at low n as already discussed by Farley *et al.* in Ref. 6.

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