Measurement of High-Angular-Momentum Fine Structure in Helium: An Experimental Test of Long-Range Electromagnetic Forces

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Fine-structure intervals separating the $n = 10$, $L = 4, 5, 6$, and 7 manifolds of helium have been measured in a fast atomic beam with precision sufficient to resolve predicted vacuumfluctuation corrections to the structure. We find the intervals (in the absence of spin) to be as follows: $10G-H$, 490.990(10) MHz; $10H-I$, 157.068(13) MHz; 10*I*-K, 60.818(10) MHz. The results set an experimental limit on the contributions of retarded long-range interactions that is comparable to their expected size.

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The study of long-range electromagnetic interactions has a long history which is largely theoretical. In 1948, Casimir and Polder showed, using nonrelativistic quantum electrodynamics, that the dominant interaction between neutral atoms at distances much larger than 137 Bohr radii was nonclassical (proportional to h) and given by¹

 $V_{CP} = (23\pi c/4\pi) \alpha_d(1) \alpha_d(2)/R^7$,

where $\alpha_d(i)$ is the dipole polarizability of the *i*th atom and R is the separation of the atoms. This interaction, when summed over the atoms composing two neutral conducting parallel plates, gives rise to the so-called "Casimir force." $2,3$ In 1978, Kelsey and Spruch showed that an analogous long-range interaction should exist between an electron and a polarizable system and be given, for large R , by⁴

$$
V_{\rm KS} = (11/4\pi) (\hbar/mc) e^2 \alpha_d/R^5.
$$
 (1)

This prediction has been recently confirmed by Feinberg and Sucher, who also gave a formal treatment of the problem with more general validity $(R \ge a_0)$ ⁵ Kelsey and Spruch showed that the potential of Eq. (1) could be interpreted as a dynamic effect of zero-point fluctuations of the electromagnetic field, $6a$ conclusion that has also been reached in the case of the macroscopic "Casimir force."² Forces of this type can be described in a number of ways.⁷⁻⁹ Their analogs in QCD are also of interest.¹⁰

Experimental studies of these long-range interactions have been confined to macroscopic tests of the "Casimir force" and closely related phenomethe "Casimir force" and closely related phenome-
na.^{11–15} The results, while consistent with the expected result, are not sufficiently precise to give a quantitative test of more than moderate $(15%)$ precision. A more precise test may be possible by observing the effect of the long-range potential of Kelsey and Spruch on the binding energies of helium Rydberg states, a system where other contributions are known very accurately.⁴ In order to make such a test, we have measured fine-structure intervals between high-angular-momentum Rydberg states of helium. We find that the long-range effects, if present, are substantially smaller than suggested by a direct application of Eq. (1).

The experimental method consisted of passing a fast atomic helium beam, which was generated by charge-exchange neutralization of a 12-keV He+ beam, through a transmission line in which rf transitions between adjacent L states in $n=10$ were driven. Detection of the transitions was accomplished by selectively exciting the lower state in the transition to $n = 27$ with a CO₂ laser (λ $=20.57 \,\mu\text{m}$) that was Doppler tuned by changing the intersection angle of the laser and atomic beams. Atoms excited to $n = 27$ were Stark ionized and the resulting ions deflected into a Channeltron detector. The apparatus is shown schematically in Fig. 1. Following charge exchange, residual ions and neutrals with $n \ge 24$ were removed from the beam with an electric field at point 1 in the figure. At point 5, $n = 10$ states were resonantly excited and then ionized at point 6 and detected. Figure 2 shows the detected ion flux as a function of beamintersection angle, and indicates the presence of high-L Rydberg states with a nearly statistical population distribution. The ion flux from the $10(L)$ state served as a detector for resonant rf transitions between $10(L)$ and $10(L+1)$ states driven at point 3 in Fig. 1. The signal strength was enhanced by depleting the natural $10(L)$ population with a

FIG. 1. Schematic drawing showing the fast atomic beam apparatus.

second resonant laser excitation to $n = 27$ applied at point 2. A typical scan of the $10H$ -I resonance, obtained in about 3 h, is shown in Fig. 3. Similar scans were obtained for the $G-H$ and I-K intervals. The latter, complicated by the lack of resolution of the $10I$ and $10K$ states in the laser excitation, was obtained by inverting the $10H$ and $10I$ populations with an rf field at point 4 and detecting the $10H$ population at point 5.

The shape of the resonances in Fig. 3 is determined by the transit time $(1.4 \mu \text{sec})$ through the rf region. The characteristic four-peak structure is due to the magnetic fine structure of the states. To a precision adequate for the present measurement, 16 it is described by

$$
H_{\text{spin}} = \alpha^2 \mathcal{R} \left(\frac{\vec{L} \cdot \vec{S}_R}{r^3} - 2 \frac{\vec{L} \cdot \vec{S}_c}{r^3} + 2 \frac{\vec{S}_R \cdot \vec{S}_c - 3(\vec{S}_R \cdot \hat{r}) (\vec{S}_c \cdot \vec{r})}{r^3} \right) + V_x \left(\frac{1}{2} + 2(\vec{S}_R \cdot \vec{S}_c) \right),\tag{2}
$$

where \overline{S}_R and \overline{S}_c are the spins of the Rydberg and core electron, and V_x is taken to be 0.025 MHz for the $10G$ state and zero for the higher L states; $\mathcal{R} = 1$ Ry. To an adequate approximation, only four transitions are allowed, each connecting corresponding spin states of adjacent manifolds.¹⁷ In the absence of the interactions represented by Eq. (2), all four components would have a common position which we call the "spinless" interval. The displace-

ment of each component from that position was calculated from Eq. (2) and the data fitted by a superposition of lines with these relative displacements to obtain a measurement of the spinless interval. The smooth curve in Fig. 3 is the result of such a fit. The fitted line centers from two such line scans

FIG. 2. Ion flux into Channeltron following laser excitation and Stark ionization vs intersection angle of laser and atomic beams. $(\theta = 0$ corresponds to parallel beams.)

FIG. 3. Ion flux synchronous with modulation of rf amplitude vs rf frequency showing 10H-I resonances.

TABLE I. Comparison of experimental measurements of $n = 10$ spinless intervals with theory. Column 2 shows the average Stark-shift correction applied to the measured intervals. Column 6 shows the estimated correction with the Kelsey-Spruch potential, which is not included in column 4. (All energies in MHz.)

Interval	$\Delta \nu$.	Expt.	Theory (Refs. 19, 23)	$Expt. - theory$	Kelsey-Spruch (Ref. 4)
$10G-10H$	0.023(9)	490.990(10)	491.02(52)	$-0.03(52)$	-0.65
$10H-10I$	0.035(12)	157.068(13)	157.058(10)	0.010(16)	-0.131
$10I-10K$	0.014(6)	60.818(10)	60.818(1)	0.000(10)	-0.033

with the direction of rf propogation chosen parallel and antiparallel to the atomic beam were averaged to give a single determination of the spinless interval. This procedure was repeated three times for each interval.

The primary systematic uncertainty in these measurements was due to the presence of stray electric fields within the rf interaction region. Since possible motional electric fields were eliminated by a double magnetic shield which gave $B \le 10$ mG, these were probably caused by charging of nominally conducting surfaces. The quadratic Stark-shift rates of the $10G-H$, $10H-I$, and $10I-K$ transitions were calculated to be $-12.1(7)$, $-15.6(6)$, and $-7.9(1.7)$ MHz/(V/cm)² respectively,¹⁷ where the uncertainties are due to variations between magnetic substates whose relative population was unknown. As a sensitive check for the presence of stray fields, the position of the three-photon $10^{-} I_6$ -10⁻ M_9 transition,¹⁸ which shifts at a much faster rate with electric field $[+132(15)]$ $MHz/(V/cm)^2$], was regularly monitored. The measured position was corrected for an ac Stark shift and then compared to the position calculated by Drachman $[101.614(1) \text{ MHz}]$.¹⁹ Any difference was assumed to be due to stray fields and was used to compute a Stark-shift correction to the measured centers of the one-photon intervals. Table I shows the average Stark-shift correction (Δv_s) applied for each measured interval.

A second systematic effect is related to the small counterpropagating wave within the rf interaction region. This small reflected wave $(V_{-}/V_{+} \le 0.04$ over the range of these measurements) will shift the apparent center of the resonance slightly. To the extent that the two ends of the rf region are physically equivalent, the average of up- and down-shifted lines should be unshifted. Any physical difference between the two ends of the rf region could bias the average result. In that case, however, similar measurements obtained with the spatial orientation of the rf region reversed within the apparatus would contain an opposite bias. Results obtained in the two orientations were found to be indistinguishable, indicating that the net shift for either orientation was small. We take the average of the results of the two orientations as the best estimate of the line center.

The final results for the $10G-H$, $10H-I$, and 10I-K intervals, shown in Table I, column 3, are much more precise than the only previous measurements.²⁰ Column 4 shows the values predicted by the calculation of Drachman,¹⁹ after correcting for relativistic effects on the dipole polarizability of the ' $He⁺ core, ^{21, 22}$ and for the second-order effects of dipole polarization.²³ The long-range effects predicted by Kelsey and Spruch, which are not included in column 5, are estimated in column 6 by calculating the expectation value of Eq. (1) in the relevant states. The differences between our measurements and the partial predictions of column 4, given in column 5, show no evidence for any corrections the size of column 6. Thus, the experimental evidence suggests that vacuum-fluctuation effects, if present, are substantially smaller than the simplest estimate. Since the Kelsey-Spruch potential is thought to be valid only asymptotically $(r \ge 137a_0)$, it gives only a crude estimate for $n = 10$ states. It was, however, the only estimate available prior to this experiment. ²⁴

The precision of the present experimental results indicates that long-range effects of this type must be included in the theory in order to give a satisfactory account of the structure. The calculations of Drachman demonstrate that, for sufficiently high L, such effects will not be obscured by uncertainties in the Coulomb energies. A detailed test, therefore, continues to seem possible in these states. Such a test will require improved experimental results, which appear quite feasible with the present technique. It will also require a more fundamental theoretical treatment of the structure of these Ryd-

berg states which consistently includes the Coulomb and long-range interactions as well as electron spin and relativistic corrections.

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24C. K. Au, G. Feinberg, and J. Sucher now calculate the expected contributions to the intervals measured here with the method of Ref. 5 and find results smaller than column 6 of Table I by about an order of magnitude and consistent with the experimental results reported here. See following Letter [Phys. Rev. Lett. 53, 1145 (1984)].