Measurement of the Shift of Rydberg Energy Levels Induced by Blackbody Radiation

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Using high-precision laser spectroscopic techniques we have measured the predicted shift of Rydberg energy levels induced by blackbody radiation. Fractional shifts of $\sim 2\times 10^{-12}$ are consistent with theoretical predictions.

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A fundamental interest in atomic physics is the interaction of atoms with the ever-present radiation fields. The atom's coupling with the vacuum modes causes the "spontaneous" decay of excited states and gives the major contribution to the Lamb shift. The possibility that real field quanta at finite temperatures give corrections to atomic level positions (and the Lamb shift specifically) has been studied for thirty years.[1] The result is that blackbody radiation (BBR) does induce temperature-dependent shifts of atomic energy levels but, for the Lamb shift and most low-lying states, the shifts are of a generally negligible magnitude.[2,3] Only in contemporary atomic frequency standards[4] do the shifts approach observability. Recently Gallagher and Cooke[5,6] noted that for high-lying Rydberg states there is a differential positive shift (though small) of all Rydberg states relative to low-lying states. We report here the first measurement of such BBR-induced atomic energy-level shifts. The other important effects of BBR on Rydberg atoms are stimulated population transfer effects,[5-8] or their absence,[9] including BBR-induced photoionization[10] and cooperative effects.[11]

Following the detailed calculations of Farley and Wing,[12] we focus on the predicted Rydberg energy-level shift. The dynamic Stark shift of the state "a" can be written as

$$\delta\nu_a = \frac{e^2}{8\pi\hbar^2}\sum_{i,b}\int_0^\infty |\langle a|r_i E_i|b\rangle|^2 [(\omega_a-\omega_b-\omega)^{-1}+(\omega_a-\omega_b+\omega)^{-1}]d\omega, \quad (1)$$

where the perturbing electric field components E_i come from the Planck radiation distribution, and where b includes both discrete and continuum states.

For highly excited Rydberg states (principal quantum number $n \gtrsim 15$), we have $|\hbar(\omega_a-\omega_b)| \ll kT$ and the shift reduces to $e^2(kT)^2/6\hbar^2mc^3$. Thus, all highly excited Rydberg states, independent of their other properties, are shifted to higher energy. The amount is +2.4 kHz at 300 K. The same value is given by the formalism of Avan *et al.*[13] which has the physical interpretation of the BBR-induced shift as an additive kinetic energy of the "nearly free" Rydberg electron. The Welton potential-averaging term[13,14] is smaller. Certain laser-induced Stark shifts can also be interpreted in this manner.[15]

To measure the BBR-induced atomic energy-level shift, we used the experimental system depicted schematically in Fig. 1. High-resolution measurements on atomic Rydberg states are naturally accomplished by use of Doppler-free two-photon spectroscopy in an atomic beam, with the resolution further enhanced by use of the two-zone Ramsey fringe method.[16] The Rb atoms travel vertically and interact successively with the laser field in two (155 μm spot radius, 1.7 mm separation) zones created by the folded optical buildup cavity [power buildup, $\times$ (50 to 80)]. BBR from an isolated and separately heated ($350 \leq T_{BB} \leq 1000$ K) blackbody source is focused on the atom/laser interaction region by a gold-coated spherical mirror (used at $f/0.78$). A mechanical chopper, mounted in an aluminum wall, is used to encode the blackbody shift information by chopping the blackbody source radiation on and off. By synchronizing the laser frequency steps with the chopping function, we simultaneously obtain two spectra where every other data point is taken with the blackbody radiation (BBR) present. dc electric fields in the interaction region are precisely controlled by a three-axis Stark electrode structure.[17] A $\sim$10-G magnetic field along the atomic beam direction separates the $5s$ to $36s$, $F=3$ transition into its eleven Zeeman components. We study the magnetic-field-independent $|M_F|=3$ to $|M_I+M_S|=3$ component.[16]

In applying the theoretical relationship of Eq. (1)

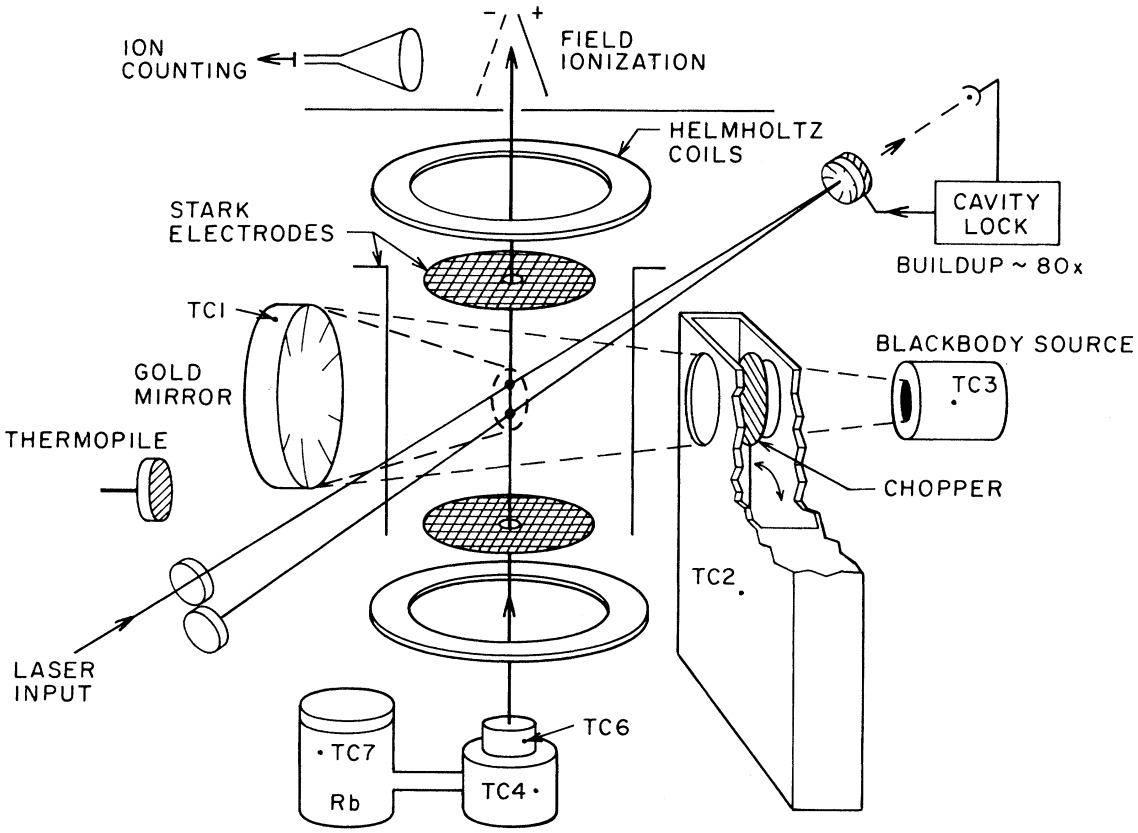

FIG. 1. Schematic of apparatus. A Rb atom in the beam (vertical arrow) may interact twice with the folded intracavity field of the "buildup" resonator located inside the vacuum. Excited Rydberg population is quenched by field ionization to yield an ion count. The blackbody thermal radiation is chopped and focused onto the laser/atom interaction region. Several thermocouples (TC) monitor relevant temperatures.

to our experimental situation, several factors need to be taken into account: the blackbody source emissivity[18,19] ($\epsilon = 0.97$), the gold mirror reflectivity (0.98), the effect of focusing, anisotropy, and polarization of the BBR, and the chopping wave form. These factors (mainly the reduced solid angle) reduce the expected shift by the factor of 0.136 $\pm$ 10%, but we compensate for this loss by raising the blackbody temperature.[18] For this experimental apparatus and large principal quantum number the theory predicts a positive (blue) frequency shift for transitions from the ground state to all highly excited Rydberg states (of all atoms) of

$$\delta\nu_c = (0.136)\frac{\alpha k^2}{6\pi mc^2}[T_{\mathrm{BB}}^2 - (353)^2]. \qquad (2)$$

Here a residual temperature of 353 K is the equilibrium temperature of the Rb beam environment. With the blackbody source temperature at 800 K this predicts an energy level shift of 1.9 kHz, corresponding to a frequency shift of 950 Hz at the laser frequency for the two-photon transition.

Because of this very small expected shift (fractional shift of $\sim 2\times 10^{-12}$) the dye-laser scan and frequency control require the highest precision possible. We rely on recently developed[19] optical heterodyne techniques for control of the laser frequency relative to a weak, but isolated, hfs component in an I_2 cell. A dye-laser linewidth of ~ 100 Hz with a drift rate of <100 Hz/(5 min) is obtained, with long-term reproducibility of <5 kHz for ~ 6 months.[19]

With the apparatus of Fig. 1 we obtain the Rydberg state population as a function of the input laser frequency; see Fig. 2. This broad scan of 1.53 MHz over the entire $|M_F| = 3$ Zeeman component shows the high contrast and narrow spectral width [40 kHz full width at half maximum (FWHM)] of the Ramsey interference fringes on the transit-limited (700 kHz FWHM) Doppler-free two-photon background signal. Shown as a solid line through the data points in Fig. 2 is the best-fitting two-photon Ramsey fringe line shape.[16,20,21] This theory includes the important physics of the Gaussian transverse nature

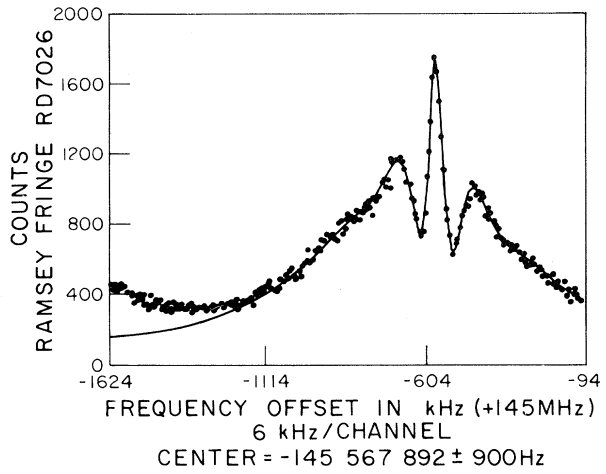

FIG. 2. Two-photon Ramsey fringe profile. Trace shows the ~700-kHz-wide single-zone transit-broadened resonance of $5s\,|M_F| = 3 \rightarrow 36s\,|M_S + M_I| = 3$ two-photon line at $\lambda = 595.429\,80$ nm. Superposed is 40-kHz-FWHM two-zone Ramsey interference resonance. Ramsey resonance shows far less laser light shift (red) than single-zone transit resonance. Other Zeeman components are just beyond limits of scan.

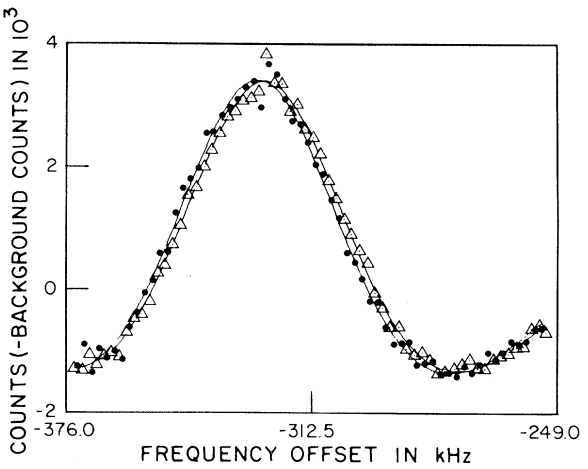

FIG. 3. Central Ramsey fringe: Chopper running with blackbody source heated to 876 K. Data taken with source exposed to BBR (triangles) show blue shift of $+1400 \pm 310$ Hz (at the laser frequency) relative to the chopper-blocked data (circles). There is no shift with the source not heated.

of the laser mode, the laser-induced light intensity shifts, the Maxwell-Boltzmann distribution of atomic velocities (and the transit broadening which the velocities imply), the second-order relativistic Doppler shift, the decay of the atomic two-photon coherence between the two atom-laser interactions, and the decay of the excited-state population before detection. The agreement between the theoretical line shape and the experimental data is remarkably good, showing nearly random residuals dominated by the counting statistics.

A shift in the position of the resonance—such as that induced by thermal radiation—will be most apparent at the steep slopes on the sides of the central Ramsey fringe. Figure 3, which focuses in on just the central Ramsey fringe, shows data taken with the chopper running and with the blackbody source heated to 876 K. The data from this file are separated into two individual data sets which are then separately fitted with the Ramsey fringe line shape. In this case we clearly see the resonance shifted to higher frequencies (to the blue) when the BBR is present. A resonance shift, at the laser frequency, of $+1.4$ kHz ± 310 Hz is measured for this data set. For a similar scan but without the blackbody source heated the difference in the resonance-fit centers gives a measured shift of -48 ± 215 Hz in laser frequency units. Thus, as expected, no shift is observed when the blackbody source is not heated.

We have measured the central Ramsey fringe resonance with various blackbody source temperatures to determine the temperature dependence of the BBR-induced energy-level shift. The results, plotted in Fig. 4, show a shift which is indeed positive and increasing with temperature. Also displayed are the theoretically predicted shift [from Eq. (2)], the best weighted–least-squares fit to the data of BBR-induced shift versus temperature squared, and the uncertainties in both of these. The measured result (whose standard uncertainty is $\sim \pm 10\%$) is $\sim 30\%$ below the theoretical prediction (with no free parameters) yet it is within the uncertainty of the prediction, $^{+10}_{-30}\%$. By far the dominant contribution to, and the cause of the asymmetry in, the theoretical uncertainty is the uncertainty (± 0.5 mm) in the position of the laser/atom interaction region with respect to the focus of the blackbody radiation. (A misalignment of the interaction region from the exact focus can only reduce the BBR intensity and hence decrease the expected shift.) The measured temperature dependence is consistent with the T^2 prediction but is not a stringent test of that functional form. In analyzing possible systematic contributions to the measurement, we have considered the effects of BBR polarization anisotropy, electric fields, BBR-induced photoionization, thermoelectric and surface poten-

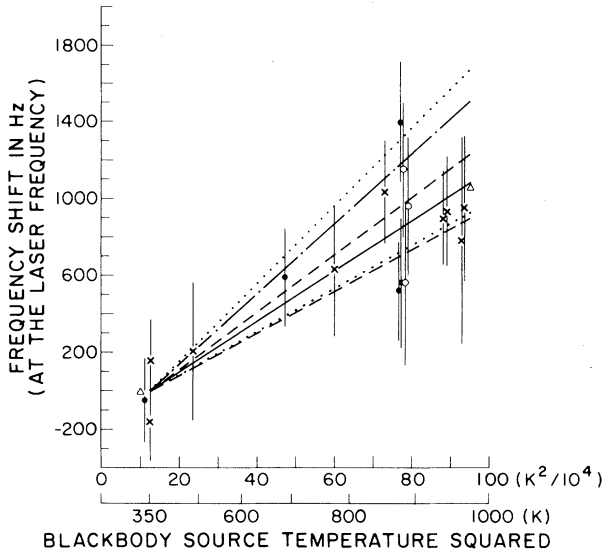

FIG. 4. Temperature dependence of measured thermal-radiation-induced energy-level shift. Points and error flags represent measured values, with best-fitting straight solid line; error limit on experimental fit denoted by dashed lines; theoretical prediction of Eqs. (1) and (2) calculated for our experimental situation (dot-dashed lines) with unsymmetrical error limits (dotted line) of $^{+10}_{-30}$%. The principal uncertainty is the location of the effective atom beam relative to the focus of the BB radiation.

tials, magnetic effects, BBR-induced reduction in Rydberg lifetime, atom emission from the BB source, and chopper heating and wave-form corrections.[19] They are all found to be of negligible importance for the data reported here.

The implementation of laser precision measurement and optical heterodyne techniques has allowed the measurement of atomic energy shifts with a precision not possible before. These measurements are consistent with the predicted finite-temperature radiative corrections to atomic energy levels induced by BBR. Even isolated single atoms are not independent of their local environment but rather are observably coupled to it via the thermal radiation field.

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