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SHIFT OF  $^2S_{1/2}$  HYPERFINE SPLITTINGS DUE TO BLACKBODY RADIATION AND ITS INFLUENCE ON FREQUENCY STANDARDS

W.M. Itano, L.L. Lewis and D.J. Wineland

*Frequency and Time Standards Group, Time and Frequency Division, National Bureau of Standards, Boulder, Colorado 80303, U.S.A.*

**Abstract.** - Frequency shifts of hyperfine splittings of  $^2S_{1/2}$  states due to the blackbody electric field are calculated. It is shown that they can be estimated from the dc hyperfine Stark shifts, which have previously been measured in the ground states of hydrogen and the alkali atoms. The shifts scale as  $T^4$ . The fractional shift for Cs at 300 K is  $-1.7 \times 10^{-14}$ , which is large enough to be significant in primary frequency standards, and should be measurable. A simple method of calculating the hyperfine Stark shifts is described, which is based on the Bates-Damgaard method for determining radial matrix elements and the Fermi-Segrè formula for determining the contact hyperfine matrix elements. It agrees with the experiment to within 12% for the entire alkali series. It is applied to  $Ba^+$  and  $Hg^+$ , for which no experimental data are yet available, and which are currently of interest for frequency standards. At 300 K, the fractional shifts are  $-9.9 \times 10^{-17}$  and  $-2.4 \times 10^{-15}$  for  $Hg^+$  and  $Ba^+$ , respectively. The shift due to the blackbody magnetic field is  $-1.3 \times 10^{-17} [T(K)/300]^2$  for any  $^2S_{1/2}$  state.

**Introduction.** - The most accurate and stable atomic frequency standards are based on hyperfine transition frequencies in  $^2S_{1/2}$  ground states, such as in  $^{133}Cs$ ,  $^1H$ , and  $^{87}Rb$ . In this communication, we estimate the temperature-dependent shift of  $^2S_{1/2}$  hyperfine splittings due to the blackbody radiation field. We note that this effect is large enough to be observable in a Cs atomic beam apparatus. The shift of the Cs hyperfine splitting at  $T = 300$  K from the unperturbed ( $T = 0$  K) value causes a frequency offset which is significant for primary frequency standards. However, since the temperature of these standards is kept very stable (for other reasons), the correction can be made very precisely.

**Blackbody Shifts.** - According to the Planck radiation law,

$$E^2(\omega)d\omega = B^2(\omega)d\omega = \frac{8}{\pi} \frac{\alpha^3}{\exp(\omega/kT) - 1} \omega^3 d\omega, \quad (1)$$

where  $E^2(\omega)d\omega$  [ $B^2(\omega)d\omega$ ] is the squared amplitude of the blackbody electric (magnetic) field in a bandwidth  $d\omega$  around  $\omega$ . [Atomic units (a.u.) are used unless otherwise specified ( $\hbar = m_e = e = 1$ )]. The mean-squared fields are

$$\begin{aligned} \langle E^2(t) \rangle &= \frac{1}{2} \int_0^{\infty} E^2(\omega) d\omega = \frac{4\pi^3}{15} \alpha^3 (kT)^4 \\ &= (8.319 \text{ V/cm})^2 \times [T(K)/300]^4, \end{aligned} \quad (2)$$

and

$$\begin{aligned} \langle B^2(t) \rangle &= \frac{1}{2} \int_0^{\infty} B^2(\omega) d\omega \\ &= (2.775 \times 10^{-6} \text{ T})^2 \times [T(K)/300]^4. \end{aligned} \quad (3)$$

Gallagher and Cooke [1] pointed out that these fields induce temperature-dependent shifts of transition frequencies in atoms and molecules through the ac Zeeman and Stark effects [2,3] They estimated the fractional blackbody ac Zeeman shift of the ground-state hyperfine splitting in H or Cs to be about  $10^{-16}$  at  $T = 300$  K.

We have derived the following expression for this shift in any  $2S_{1/2}$  ground state, which is valid at zero dc magnetic field and at temperatures such that the peak of the blackbody spectrum is at a much higher frequency than the hyperfine frequency. We find

$$\begin{aligned} \delta\omega_{\text{hfs}}/\omega_{\text{hfs}} &\cong -\frac{\alpha^2}{24} (g_J - g_I)^2 \int_0^{\infty} \frac{B^2(\omega)}{\omega^2} d\omega \\ &= -(\pi/18)(g_J - g_I)^2 \alpha^5 (kT)^2 \\ &\cong -1.304 \times 10^{-17} [T(K)/300]^2. \end{aligned} \quad (4)$$

where  $g_J$  and  $g_I$  are the electronic and nuclear  $g$  factors, respectively. In the last line, we have assumed that  $g_J = 2$  and that  $|g_I/g_J| \ll 1$ .

At laboratory temperatures, the blackbody ac Stark shift of the hyperfine splitting, which has previously been neglected, is generally larger than the ac Zeeman shift. The ac hyperfine Stark shift due to an electric field of frequency,  $\omega$ , is approximately equal to the dc hyperfine Stark shift due to a static field with the same rms value, if  $\omega \ll \omega_{\text{res}}$ , where  $\omega_{\text{res}}$  is the lowest allowed electric dipole transition frequency. The correction is of order  $(\omega/\omega_{\text{res}})^2$ . For

the ground states of any of the alkali atoms,  $(w/w_{\text{res}})^2 \lesssim 3 \times 10^{-3}$ , where  $w$  is the frequency corresponding to the peak of the blackbody spectrum at 300 K. Therefore, at 300 K, the blackbody ac hyperfine shift is approximately equal to the shift caused by a dc field of 8.3 V/cm.

The dc hyperfine Stark shift was first observed in Cs by Haun and Zacharias [4]. Later, it was observed in H (see Ref. 5) and other alkali atoms [6]. These experiments measured the Stark shift of the  $(F = I + \frac{1}{2}, M_F = 0) \leftrightarrow (F = I - \frac{1}{2}, M_F = 0)$  transition. They can be considered to be measurements of the scalar hyperfine polarizabilities, which are independent of  $M_F$  and the orientation of the electric field, since the contributions from the tensor polarizabilities can be estimated and are less than the experimental uncertainties [7]. Only the scalar polarizability contributes to the blackbody ac Stark shift, because of the isotropy of the blackbody radiation. The fractional ac Stark shift of the Cs hyperfine splitting can be estimated from the measured dc hyperfine polarizability (see Table 1 of Ref. 6) and Eq. (2) to be  $-1.69(4) \times 10^{-14} [T(K)/300]^4$ . This shift is large enough to affect the calibration of primary Cs frequency standards and therefore should be taken into account. For example, the fractional uncertainty of one primary Cs frequency standard (CS 1 of the Physikalisch-Technische Bundesanstalt) is stated to be  $6.5 \times 10^{-15}$ , but has not been corrected for the blackbody shift [8]. For H and Rb, the shifts are too small to be experimentally significant at present.

The theory of the dc hyperfine Stark shift of  $2S_{\frac{1}{2}}$  ground states is quite well developed, and the calculations are in good agreement with the experiments. For hydrogenic atoms and ions, an analytic solution has been obtained [9,10]. Numerical calculations have been made for the neutral alkali atoms [11-15].

We write the third-order perturbation expression for the scalar fractional dc hyperfine Stark shift of the  $ns \ 2S_{\frac{1}{2}}$  state of an alkali-like atom or ion in the following form, which is independent of the spin and magnetic moment of the nucleus:

$$\delta w_{\text{hfs}}/w_{\text{hfs}} = -kE^2 = -(k_1 + k_2)E^2, \quad (5)$$

where

$$k_1 = \sum_{n''J} \frac{(n \ 2S_{\frac{1}{2}} || r || n'' \ 2P_J)^2}{6[W(n''PJ) - W(nS)]^2} \quad (6a)$$

and

$$k_2 = - \sum_{n'_J, n' \neq n} \frac{(n \ 2S_{\frac{1}{2}} || r || n'' \ 2P_J)}{3[W(n''PJ) - W(nS)]} \frac{(n' \ 2S_{\frac{1}{2}} || r || n'' \ 2P_J) \psi_{n's}(0)}{[W(n's) - W(nS)] \psi_{ns}(0)}. \quad (6b)$$

The reduced matrix elements of  $\vec{r}$ , the position operator of the valence electron, are defined with the conventions of Edmonds [16]. We choose the phases of the radial wave functions so that they are real.  $W(n^1P_J)$  and  $W(nS)$  are the energies (not including the hyperfine interaction) of the  $n^1p \ ^2P_J$  and the  $ns \ ^2S_{1/2}$  states, respectively, and  $\psi_{ns}(0)$  is the value of the  $ns \ ^2S_{1/2}$  wave function at the origin. The dc magnetic field is assumed to be so small that the Zeeman splitting is much less than the hyperfine splitting.

We have developed a simple method of approximately evaluating Eqs. (6a) and (6b). We calculate the radial matrix elements using the Coulomb (Bates-Damgaard) approximation [17] and the values of the s-state wave functions at the origin using the Fermi-Segrè formula [18]. We have used this method to calculate the scalar fractional ground-state hyperfine polarizabilities of Li, Na, K, Rb, and Cs and have obtained agreement with experiment to within 12% or better in all cases. The lowest three p states and the lowest five s states were included in the basis.

This method can be used for other atoms, for which no experimental data or calculations have yet been published, such as the singly ionized alkaline earths. We have carried out the calculations for the ground states of  $Hg^+$  and  $Ba^+$ , which are currently of interest for applications in stored-ion frequency standards [19,20]. In atomic units,  $k = 37.9$  for  $Hg^+$  and  $k = 902$  for  $Ba^+$ . The conversion between atomic and laboratory units of  $E^2$  is given by

$$E^2(\text{a.u.}) = 3.782 \times 10^{-20} E^2 [(\text{V/cm})^2]. \quad (7)$$

At 300 K, the fractional blackbody ac hyperfine Stark Shifts are  $-9.9 \times 10^{-17}$  and  $-2.4 \times 10^{-15}$  for  $Hg^+$  and  $Ba^+$ , respectively. In rf trap experiments, the ac Stark shift due to the trapping fields may be larger.

The ground-state hyperfine shift due to an ac electric field of magnitude  $E(t) = E(\omega) \cos \omega t$  can be obtained by the same method that was used to derive Eqs. (6a) and (6b), except that the formula for the ac Stark shift [2,3] is used. For Cs at 300 K, the blackbody ac Stark shift is 1.4% greater than the shift due to a dc field of the same rms value.

The blackbody shift could be observed in a Cs frequency standard which was modified so that the temperature of a tube surrounding the atoms in the resonance region could be varied. If the temperature were changed, for example, from 300 K to 400 K, the fractional frequency shift would be  $3.7 \times 10^{-14}$ . If the frequency standard had the same frequency stability as NBS-6, the primary frequency standard of the United States, [21] this shift could be determined to 30% or better in an averaging time of several hours.

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