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COMMENTS AND ADDENDA

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Quadratic Stark Shifts of Zeeman Levels in Alkali Atoms

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An atomic-beam magnetic-resonance apparatus was used to measure the difference in the quadratic Stark shifts of Zeeman levels in the ground states of Na and K. The results of this work are reported. Also, previously reported related work is discussed.

This addendum is a supplement to the work previously reported by this laboratory on the difference in the quadratic Stark shifts of the Zeeman levels $(F = I + \frac{1}{2}, m_F = -I - \frac{1}{2})$ and $(I + \frac{1}{2}, -I + \frac{1}{2})$ in the ground states of alkali atoms.^{1,2}

A resonance shift, quadratic in the applied electric field, of -127 ± 20 cps at 10^5 V/cm for Cs was reported in Ref. 1. The experiments were performed on an atomic-beam magnetic-resonance apparatus. The transition was induced in a Ramsey double-hairpin structure located in the uniform residual field (~10 G) of an electromagnet. A pair of parallel, metal electric field plates were located in the region between the rf loops. To study the quadratic Stark effect, large ac (frequency ω) and dc voltages were applied to the field plates. The signal at the detector, which corresponded to the point of maximum slope on either side of the central peak of the Ramsey pattern, contained components modulated at ω (dc \neq 0) and 2ω because of the quadratic Stark interaction. Phase-sensitive detection techniques were used to separate out these components. Modulation at ω and 2ω of the Zeeman levels by a magnetic dipole interaction was likely to have occurred in these experiments. This magnetic effect was believed to have arisen from an undetermined motion of the uniform-field magnet pole pieces. The electric-field-plate structure was sandwiched between these pole pieces. Electrostrictive effects in the insulators

of the plate structure or electrostatic attraction between the plates and the pole pieces probably caused the motion. The 20-cps uncertainty was adopted to cover this possibility.

Because of the possibility of this magnetic effect, the electromagnet was replaced by a Helmholtz structure and new electric field plates. Also, the ac-modulation scheme was replaced by a switched-dc system. The new experiments were conducted on two apparatuses which differed in the lengths of their resonance regions. Measurements for Na and K using the shorter apparatus are reported here. More recent work using the longer apparatus is reported elsewhere.²

The new C-field structure incorporated two pairs of rectangular Helmholtz coils mounted at right angles to each other and enveloping the electric field plates and rf loops. One set of the Helmholtz coils, the H_z coils, produced a horizontal magnetic field \bar{H}_{z} aligned with the electric field \bar{E} to within a degree. The other set, the H_v coils, produced a vertical field \vec{H}_{v} . The resultant field $\vec{H} = \vec{H}_{y} + \vec{H}_{z}$ could be rotated with respect to \vec{E} by changing the current in the H_{ν} coils. The current in the H_{v} coils was adjusted so that the resultant C field was within 0.1° of being parallel to the E field and was roughly parallel to the deflecting fields of the A and B magnets. The strength of the C field was ~ 1 G. The rf loop separation of the short apparatus was 26.4 cm. The length of the

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electric field plates situated between the rf loops was 12.7 cm. The gap between the plates was 0.190 ± 0.0025 cm. The entire resonance region was magnetically shielded.

Digital signal-processing techniques were used. A high voltage applied to one of the field plates was periodically switched on and off. The resonance signal at the detector was accumulated in two counters (one for the E field on and the other for the E field off) which were gated synchronously with the switching frequency. A steady field during the counting time was achieved by introducing the appropriate delays into the counting cycle.

The effect of the motional magnetic field $[(\vec{v}/c) \times \vec{E}]$ was minimized by adjusting the current through the H_y coils so that the resonance shift linear in $|\vec{E}|$ was zero.³ The details of the apparatus and switched-dc techniques are discussed elsewhere.^{4,5}

Figure 1 shows the shift δf in the resonance as a function of E^2 for Na and K. Results are summarized in Table I where the constant k is defined by $\delta f = kE^2$. The data have been modified by the "filling factor" correction which takes into account



FIG. 1. Resonance shift δf versus E^2 for Na and K. This data was taken on the short Helmholtz C-field apparatus. The data are corrected for the filling factor.

TABLE I. Summary of quadratic Stark-effect results.

	k^{a} (10 ⁻¹⁰ cps/	k^{b} (10 ⁻¹⁰ cps/	k^{c} (10 ⁻¹⁰ cps/	k^{d} (10 ⁻¹⁰ cms/
Element	V^2/cm^2	V^2/cm^2)	V^2/cm^2)	V^2/cm^2
Cs	•••	-137.2	- 155	-126.8
Rb ⁸⁷	•••	± 7.9 -104.4	- 150	± 8.7
101-85		±6.7		±10.9°
Rb	•••	-22.8 ± 1.2	- 39	-15.6 ± 2.0
Na	-9.7 ± 1.9	-11.18	- 13	+5.2
К	-5.0 ± 1.5	± 0.96 - 4.8	- 7	± 1.8
		± 0.31		± 4.0

^aObtained with the short Helmholtz structure. ^bObtained from Ref. 2.

^cTheoretical.

^dObtained from Ref. 1. Also, J. P. Carrico, Ph.D. thesis, Brandeis University, 1966 (unpublished).

⁶Owing to a typographical mistake, the value of k originally reported in Ref. 1 for Rb⁸⁷ was in error.

the fact that the electric field plates extend only 48% of the distance between the centers of the rf loops.⁶ The quoted errors are three times the standard deviation of the mean plus 10% for systematic effects (uncertainties in the filling factor, calibration of the high voltages, and separation of the electric field plates). Although not as precise, the results agree within their stated errors with those reported in Ref. 2.

The values of k reported in Ref. 1 and in Ref. 2 are also included in Table I. The quoted errors for the Ref. 1 data are three times the standard deviation of the mean plus 5% for uncertainties in the filling factor and the separation of the field plates (the possibility of a systematic magnetic effect has been left out). The quoted errors for the data of Ref. 2 are three standard deviations of plus 2% for systematic effects.

We compared the Na and Rb⁸⁵ results to see if a magnetic effect could possibly account for the difference for each element between the data of Ref. 1 and 2. (Only the Na and Rb⁸⁵ data could be compared because of the poor result for K in Ref. 1, and the data of Ref. 1 and 2 agreed within their stated errors for Cs and Rb⁸⁷.) These differences normalized against the appropriate g_F values are

 $\Delta_n k(\text{Na}) = (-33 \pm 4) \times 10^{-10} \text{ cps/V}^2/\text{cm}^2$

and
$$\Delta_n k(\text{Rb}^{85}) = (-22 \pm 8) \times 10^{-10} \text{ cps/V}^2/\text{cm}^2$$
,

where the quoted errors are three standard deviations of the mean plus the estimated systematic errors. The agreement of these two normalized differences within their stated errors indicates that a systematic magnetic effect may have been mainly responsible for the differences between the two sets of data. The motion of the uniform-magnet pole pieces in the experiments of Ref. 1 would be a likely candidate for this effect.

The rf power shift reported in Ref. 2 was not observed in the experiments of Ref. 1. This shift was attributed to a multiple-quantum-transition effect. A possible explanation for its absence in the Ref. 1 work is that the uniform-magnetic field strength was ~ 10 G, whereas ~ 1 G was used in the experiments reported here and in Ref. 2. The conditions for multiple quantum transitions are expected to be more favorable as the Zeemanlevel separations are decreased. This expectation is supported by the observation in Ref. 2 that the power shift depended on the strength of the uniform magnetic field.

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Theories of Collective Motion in Liquids

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The concept of collective modes of a system is important both for the theoretical interpretation of scattering experiments and in the calculation of transport coefficients from microscopic dynamics. The frequencies and damping of these variables correspond to the experimental neutron energy peaks and also occur in any theory of thermal conductivity of a liquid. Below we compare some recent approaches to the problem, consider criteria for collective variables, and discuss applications.

The essence of Zwanzig's¹⁻⁴ approach is the following. One considers a classical system described by the Liouville equation. The collective variables of the system are determined by a variational principle, and are solutions of the eigenvalue problem for the Liouville operator. The frequencies are eigenvalues of a matrix whose elements are correlation functions of the trial functions, and are those obtained from the macroscopic equations of hydrodynamics.

Mori's theory^{5,6} is applicable to both classical and quantum systems, and its generality allows discussion of the validity of the variational method as follows. The question is: In what respect do the variational eigenvalues correspond to the true normal-mode oscillations of the system, or, when is a set of functions a set of good collective variables ?

Let $D_k(t)$ be the normalized time- and wavevector-dependent correlation matrix ${}^6 [D_k(t)]_{ij}$ = $(A_i(t), A_j^*)$, where the $A_i(t)$ can be chosen as the number density, momentum density, and energy density fluctuations as well as the time derivatives of these. In the classical limit the relaxation functions (A, B^*) (B^* is complex conjugate of B) reduce to $\langle AB \rangle_{av}/k_BT$, where $\langle \cdot \cdot \cdot \rangle_{av}$ is an average with respect to an equilibrium ensemble. They are otherwise given in terms of a Kubo transform, 6 which is, in fact, the generalization of the