Hyperfine Structure and g_J Value of the $^4F_{9/2}$ and $^4P_{5/2}$ Levels of Cu^{63} and Cu^{65}

Arthur G. Blachman, * Donald A. Landman, † and Allen Lurio
IBM *Watson Laboratory*, *Columbia University*, *New York*, *New York* 10025
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The hyperfine structure of the $(3d^9 4s4p)$ $^4F_{9/2}$ and $^4P_{5/2}$ metastable levels in Cu^{63} and Cu^{65} have been measured by the atomic-beam magnetic-resonance method. When corrected for second-order interactions with neighboring fine-structure levels from the $(3d^9 4s4p)$ configuration, the hfs interaction constants are

$A^{63}(^4F_{9/2}) = 1323.891(1)$ MHz, $A^{65}(^4F_{9/2}) = 1418.123(2)$ MHz,
$A^{63}(^4P_{5/2}) = 2031.239(1)$ MHz, $A^{65}(^4P_{5/2}) = 2175.811(2)$ MHz,
$B^{63}(^4F_{9/2}) = 137.874(5)$ MHz, $B^{65}(^4F_{9/2}) = 127.586(19)$ MHz,
$B^{63}(^4P_{5/2}) = 78.993(4)$ MHz, $B^{65}(^4P_{5/2}) = 73.111(9)$ MHz,

The octupole interaction constants are of the order of the experimental uncertainties. From the Zeeman effect of the hfs, we find $g_J(^4F_{9/2}) = 1.3340(2)$ and $g_J(^4P_{5/2}) = 1.600(2)$. By making use of the ratio A^{63}/A^{65} in the $^2S_{1/2}$, the $^4F_{9/2}$, and the $^4P_{5/2}$ states, we infer that $g_I^{63}/g_I^{65} = 0.933\,524(5)$ and that $^{63}\Delta^{65}(^2S_{1/2}) = 0.000\,048(5)$. We also find that $Q^{63}/Q^{65} = 1.080\,54(14)$.

I. INTRODUCTION

This is the third in a series of papers[1] devoted to the study of the hfs of various metastable levels of the stable isotopes of the group IB elements: $Cu^{63, 65}$, $Ag^{107, 109}$, and Au^{197}. The measurements were made by the atomic-beam magnetic-resonance method. In this paper we consider the $(3d^9 4s4p)^4F_{9/2}$ and $^4P_{5/2}$ levels of the copper isotopes, both of which have $I = \frac{3}{2}$. Figure 1 shows the low-lying energy levels of Cu I.

We cannot observe the lowest metastable levels of copper, the $(3d^9 4s^2)^2D_{3/2,\,5/2}$ levels, because their excitation energy above the ground state is only about 1.3 eV while our detection technique requires an energy difference of about 1.8 eV.

The $3d^9 4s4p$ configuration of copper has been studied much more extensively than the equivalent levels in Au and Ag. All its levels have been identified. Landman, Levin, and Lurio[2] (LLL) have obtained wave functions for the levels of this configuration, and have given a detailed analysis of the hfs of this configuration. Here we will make use of this analysis in interpreting our results.

II. APPARATUS

The apparatus used, except for the detection system, was essentially the same as that described in the previous papers.[1,3] In brief, the metastable atoms in the beam were produced by cross electron bombardment of the ground-state beam and detected by causing the refocused beam to strike a Cs-coated surface and then collecting the electrons produced by their resulting Auger de-excitation. The electron bombardment heated source oven was run at ~1550°C.

In making a sensitive detector surface, the cesium coating is produced by a spray from a small cylindrical oven located near the surface. The oven is supported by a thin-wall stainless-steel tube connected to a separate flange mounted on the detector flange. The cesium for this oven

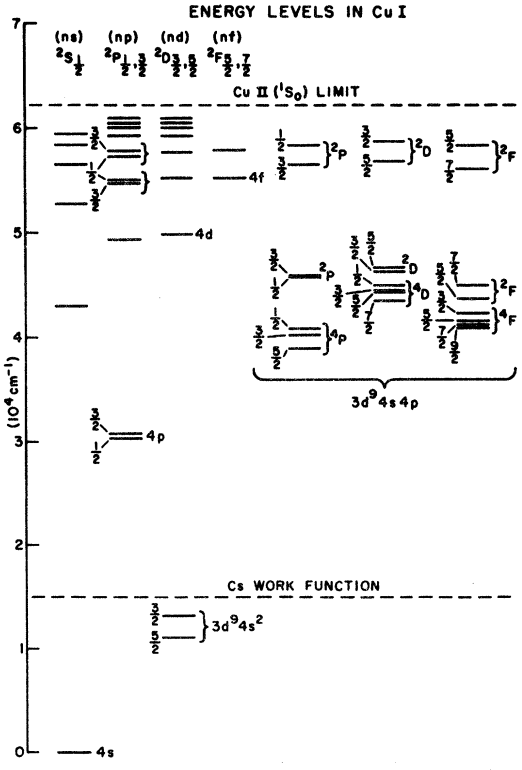

FIG. 1. Low-lying energy levels of Cu I.

is stored in sealed-off cylindrical glass ampoules which have been filled by transfer from a large ampoule under vacuum. The last step in closing the apparatus is the loading of the oven as follows: The apparatus is rough pumped and back filled with nitrogen gas and the cesium oven is removed. A cesium ampoule, cooled in liquid nitrogen, is cracked in two and the part containing cesium is loaded into the oven. The oven is then put into the apparatus which is quickly evacuated. The cesium does not react appreciably with air during the procedure which takes about a minute or less.

The detector for the Auger emitted electrons is a Bendix M-306 magnetic multiplier. We have found this multiplier to have a stable gain and low noise (1–2 pulses/sec) over many years with repeated exposures to air. We have occasionally cleaned it with procedures recommended by Bendix, but more for periodic maintenance than necessity. For detecting high-energy metastables such as the noble-gas $^3P_2$ states, the metastables are caused to strike the tungsten cathode surface and Auger de-excite. To detect low-energy metastables one needs a Cs-coated surface which must be located at the cathode position, since the magnetic field of the multiplier makes it very difficult to get external electrons into the dynode region. We therefore replaced the tungsten cathode with a brass surface at the same location, but connected to a liquid nitrogen trap by a sapphire rod. Cs was sprayed onto this surface, care being taken to prevent any direct spray from reaching the dynodes.

III. EXPERIMENTAL RESULTS

The $(3d^9 4s4p)^4F_{9/2}$ and $^4P_{5/2}$ metastable components of the beam were identified by comparing the ratios of the observed $\Delta F = 0$, $|\Delta m_F| = 1$ low-field Zeeman transition frequencies with those predicted theoretically.

In Fig. 2 we give a schematic energy-level diagram for the $^4P_{5/2}$ hfs in an applied magnetic field. A similar diagram for the $^4F_{9/2}$ state has already been given in the second paper of Ref. 1. The procedure for search and measurement of the low-field hfs transitions was the same as that

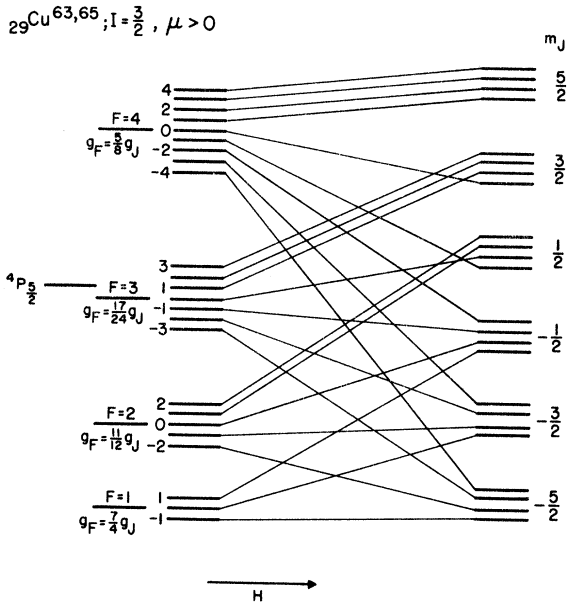

FIG. 2. A schematic plot of the magnetic field dependence of the hfs of the $^4P_{5/2}$ level.

described in the previous papers. The results are given in Table I. We were unable to observe the Cu^{65} $\Delta\nu[^4F_{9/2}; (6,m) \leftrightarrow (5,m')]$ transitions even with a beam of separated Cu^{65}, probably due to insufficient klystron power. After making the small field-dependent corrections for each line, we obtain the values for the hfs separations shown in Table II. The error quoted in each of these (and subsequent) results is three times the standard deviation of the mean.

Using these hfs intervals to analyze the observed intermediate-field Zeeman transitions, we obtain the values $g_J(^4F_{9/2}) = 1.3340(2)$ and $g_J(^4P_{5/2}) = 1.600(2)$. The g_J factor of the $^4F_{9/2}$ level is in excellent agreement with the predicted value of 1.3341 (neglecting configuration mixing). The g_J factor of the $^4P_{5/2}$ level also agrees perfectly with the intermediate coupling value of 1.599 which was calculated using the wave functions presented in LLL.

A lower limit of ~1 msec is obtained for the lifetimes of both the $^4F_{9/2}$ and $^4P_{5/2}$ levels from the transit time of the beam down the apparatus.

IV. DISCUSSION OF RESULTS

A. The hfs Interaction Constants

The zero-field hfs separations can be written to second order (for either isotope) as

$$\Delta\nu(^4F_{9/2}; F=6 \leftarrow F=5) = 6A(^4F_{9/2}) + \tfrac{2}{3}B(^4F_{9/2}) + \tfrac{16}{3}C(^4F_{9/2}) + h^{-1}[W_6{}^{(2)}(^4F_{9/2}) - W_5{}^{(2)}(^4F_{9/2})],$$

$$\Delta\nu(^4F_{9/2}; F=5 \leftarrow F=4) = 5A(^4F_{9/2}) - \tfrac{5}{24}B(^4F_{9/2}) - \tfrac{65}{6}C(^4F_{9/2}) + h^{-1}[W_5{}^{(2)}(^4F_{9/2}) - W_4{}^{(2)}(^4F_{9/2})],$$

$$\Delta\nu(^4F_{9/2}; F=4 \leftarrow F=3) = 4A(^4F_{9/2}) - \tfrac{2}{3}B(^4F_{9/2}) + \tfrac{208}{21}C(^4F_{9/2}) + h^{-1}[W_4{}^{(2)}(^4F_{9/2}) - W_3{}^{(2)}(^4F_{9/2})],$$

$$\Delta\nu(^4P_{5/2}; F=4 \leftrightarrow F=3) = 4A(^4P_{5/2}) + \tfrac{4}{5}B(^4P_{5/2}) + \tfrac{32}{5}C(^4P_{5/2}) + h^{-1}[W_4{}^{(2)}(^4P_{5/2}) - W_3{}^{(2)}(^4P_{5/2})],$$

$$\Delta\nu(^4P_{5/2}; F=3 \leftrightarrow F=2) = 3A(^4P_{5/2}) - \tfrac{9}{20}B(^4P_{5/2}) - \tfrac{81}{5}C(^4P_{5/2}) + h^{-1}[W_3{}^{(2)}(^4P_{5/2}) - W_2{}^{(2)}(^4P_{5/2})],$$

$$\Delta\nu(^4P_{5/2}; F=2 \leftrightarrow F=1) = 2A(^4P_{5/2}) - \tfrac{4}{5}B(^4P_{5/2}) + \tfrac{96}{5}C(^4P_{5/2}) + h^{-1}[W_2{}^{(2)}(^4P_{5/2}) - W_1{}^{(2)}(^4P_{5/2})],$$

where A, B, and C are the magnetic dipole, electric quadrupole, and magnetic octupole hfs interaction constants, and $W_F{}^{(2)}$ is the second-order contribution to the energy due to states with angular momentum F arising from other fine-structure levels. Neglecting the second-order terms we get the uncorrected values shown in Table III. The values $A(^4F_{9/2})^{63} = 1270$ MHz and $A(^4P_{5/2})^{63} = 2010$ MHz were obtained previously by optical measurements.[4]

TABLE I. Experimental low-field hfs data. (All units are MHz).

Level	Isotope	Transition	Frequency	$\mu_0 H$
$^4F_{9/2}$	65	$(6, m) \leftrightarrow (5, m')$	not observed	
$^4F_{9/2}$	65	$(5, m) \leftrightarrow (4, m)$	7064.184	a
			7064.199	
			7064.166	
			7064.193	
			7064.190	
			7064.190	
			7064.190	
			7064.190	
			7064.193	
$^4F_{9/2}$	65	$(4, 0) \leftrightarrow (3, 0)$	5586.607	0.46
			5586.605	0.46
			5586.604	0.46
			5586.595	0.86
			5586.601	0.86
			5586.600	4.16
			5586.602	1.95
$^4F_{9/2}$	63	$(6, m) \leftrightarrow (5, m)$	8036.898	a
			8036.894	
			8036.898	
			8036.898	
			8036.894	
			8036.908	
$^4F_{9/2}$	63	$(5, m) \leftrightarrow (4, m)$	6590.872	a
			6590.874	
			6590.870	
			6590.880	
			6590.874	
			6590.869	
			6590.880	
			6590.880	
$^4F_{9/2}$	63	$(4, m) \leftrightarrow (3, m)$	5202.922	a
		$(4, 0) \leftrightarrow (3, 0)$	5202.916	0.77
			5202.920	0.77
			5202.920	0.77
			5202.922	0.77
		$(4, -1) \leftrightarrow (3, -1)$	5203.285	0.77
		$(4, 1) \leftrightarrow (3, 1)$	5202.559	0.77
		$(4, -1) \leftrightarrow (3, -1)$	5203.281	0.77
		$(4, 1) \leftrightarrow (3, 1)$	5202.570	0.77

TABLE I. (*Cont.*)

$^4P_{5/2}$	65	$(4,m) \leftrightarrow (3,m)$	8761.974	a
			8761.982	
			8761.999	
			8761.985	
			8761.994	
			8761.994	
$^4P_{5/2}$	65	$(3,0) \leftrightarrow (2,0)$	6494.512	2.53
			6494.507	2.20
			6494.504	1.08
$^4P_{5/2}$	65	$(2,0) \leftrightarrow (1,0)$	4293.015	3.77
			4293.016	3.77
$^4P_{5/2}$	63	$(4,0) \leftrightarrow (3.0)$	8188.365	2.00
			8188.370	2.00
			8188.371	2.00
			8188.373	1.37
$^4P_{5/2}$	63	$(3,0) \leftrightarrow (2,0)$	6058.150	2.45
			6058.153	2.67
			6058.150	1.28
			6058.153	1.86
			6058.149	1.86
$^4P_{5/2}$	63	$(2,0) \leftrightarrow (1,0)$	3999.163	1.23
			3999.170	1.47
			3999.173	1.47
		$(2,1) \leftrightarrow (1,1)$	3997.405	1.32
		$(2,-1) \leftrightarrow (1,-1)$	4000.937	1.32

[a]Lines overlapped in very small field.

TABLE II. Zero-field hfs intervals. (All units are MHz.)

Level	Isotope	F	$\Delta\nu[F \leftrightarrow (F-1)]$
$^4F_{9/2}$	65	6	not observed
$^4F_{9/2}$	65	5	7064.188(9)
$^4F_{9/2}$	65	4	5586.601(6)
$^4F_{9/2}$	63	6	8036.898(6)
$^4F_{9/2}$	63	5	6590.875(6)
$^4F_{9/2}$	63	4	5202.921(3)
$^4P_{5/2}$	65	4	8761.988(12)
$^4P_{5/2}$	65	3	6494.508(6)
$^4P_{5/2}$	65	2	4293.003(3)
$^4P_{5/2}$	63	4	8188.370(6)
$^4P_{5/2}$	63	3	6058.151(3)
$^4P_{5/2}$	63	2	3999.168(6)

TABLE III. The hfs interaction constants. (All units are MHz.)

	Uncorrected	Corrected
$A(^4F_{9/2})^{65}$	1418.233(2) $+\frac{26}{7}C(^4F_{9/2})^{65}_{\text{uncorr}}$	1418.123(2) $+\frac{26}{7}C(^4F_{9/2})^{65}_{\text{corr}}$
$B(^4F_{9/2})^{65}$	129.499(19) $+\frac{260}{7}C(^4F_{9/2})^{65}_{\text{uncorr}}$	127.586(19) $+\frac{260}{7}C(^4F_{9/2})^{65}_{\text{corr}}$
$C(^4F_{9/2})^{65}$	$C(^4F_{9/2})^{65}_{\text{uncorr}}$	$C(^4F_{9/2})^{65}_{\text{corr}}$
$A(^4F_{9/2})^{63}$	1323.984(1)	1323.891(1)
$B(^4F_{9/2})^{63}$	139.501(5)	137.874(5)
$C(^4F_{9/2})^{63}$	−0.001 52(35)	−0.000 34(35)
$A(^4P_{5/2})^{65}$	2175.833(2)	2175.811(2)
$B(^4P_{5/2})^{65}$	73.322(9)	73.111(9)
$C(^4P_{5/2})^{65}$	−0.000 26(32)	0.000 01(32)
$A(^4P_{5/2})^{63}$	2031.258(1)	2031.239(1)
$B(^4P_{5/2})^{63}$	79.176(4)	78.993(4)
$C(^4P_{5/2})^{63}$	−0.000 38(18)	−0.000 10(18)

The second-order energy terms can be calculated from a knowledge of the electronic coupling in the $(3d^9 4s4p)$ configuration together with the individual electron interaction constants for the configuration.[2,4] (We neglect interconfiguration contributions to $W_F^{(2)}$.) This information is provided by the analysis of the $(3d^9 4s4p)$ configuration presented in LLL, where the following values for Cu^{63} are given:

$$a_s = 7250 \text{ MHz}, \quad a'_p = 181 \text{ MHz}, \quad a'_d = 795 \text{ MHz}, \quad b'_p = -63 \text{ MHz}, \quad \text{and } b'_d = -201 \text{ MHz}.$$

The corresponding values for Cu^{65} can be obtained by multiplying the a constants by $\mu^{65}/\mu^{63} = 1.0712(2)$,[5] and the b constants by $Q^{65}/Q^{63} = 0.9254(3)$,[6] where μ and Q denote the nuclear magnetic dipole and electric quadrupole moments, respectively. The calculated second-order corrections are shown in Table IV; the resulting corrected hfs interaction constants are shown in Table III.

The principal contribution to the second-order corrections of the $^4F_{9/2}$ level comes from the $^4F_{7/2}$ level and a small amount from each of the $(^3P)^2F_{7/2}$ and the $(^1P)^2F_{7/2}$ levels. The second-order corrections to the $^4P_{5/2}$ level are much smaller and arise principally from the $^4P_{3/2}$, $(^3P)^2D_{3/2}$, $(^3P)^2P_{3/2}$, and the $(^1P)^2P_{3/2}$ levels. (See Table V.)

TABLE IV. Second-order hfs corrections. (All units are MHz.)

	Cu^{65}	Cu^{63}
$W_6^{(2)}(^4F_{9/2})$	0	0
$W_5^{(2)}(^4F_{9/2})$	−1.879	−1.635
$W_4^{(2)}(^4F_{9/2})$	−2.033	−1.773
$W_3^{(2)}(^4F_{9/2})$	−1.200	−1.048
$W_4^{(2)}(^4P_{5/2})$	−0.015	−0.013
$W_3^{(2)}(^4P_{5/2})$	−0.268	−0.233
$W_2^{(2)}(^4P_{5/2})$	−0.242	−0.211
$W_1^{(2)}(^4P_{5/2})$	−0.111	−0.097

B. The Nuclear Quadrupole and Octupole Moments

From the results for the $^4F_{9/2}$ and $^4P_{5/2}$ levels we obtain

$$\frac{Q^{63}}{Q^{65}} = \begin{cases} \dfrac{B(^4F_{9/2})^{63}\text{corr}}{B(^4F_{9/2})^{65}\text{corr}} = 1.080\,64(16) \\ \dfrac{B(^4P_{5/2})^{63}\text{corr}}{B(^4P_{5/2})^{65}\text{corr}} = 1.080\,45(14) \end{cases}$$

in good agreement with the previously measured value of 1.0806(3).[6] The fact that the uncorrected B constants give a ratio 1.077 and 1.0798(1), respectively, is evidence of the reliability of the second-order corrections. (In this calculation we have ignored the C correction Table III which is negligible.)

The corrected values of the octupole coupling constants are too small to reliably derive either individual values or the ratio of the octupole moments of the two isotopes.

C. The hfs Anomaly and the g_I Ratio

The hfs anomaly Δ is defined by the relation

$$^{63}\Delta^{65} = (A^{65}g_I{}^{63}/A^{63}g_I{}^{65}) - 1.$$

Since Δ is very small in copper and the g_I ratio[7] is known only to a precision of 1 in 10^4, we cannot obtain Δ from the above expression. (See Table VI.) Both the g_I ratio and Δ_s, the anomaly for an s electron, can be obtained, however, by making use of the very precise A^{63}/A^{65} ratios in the ground $^2S_{1/2}$ and the metastable $^4F_{9/2}$ and $^4P_{5/2}$ states. We write for the dipole coupling constant of any level, $A = \alpha_s + \alpha$ when α_s is the s electron contribution to the hfs constant A and α is the p plus d electron contribution ($\alpha = 0$ for the $^2S_{1/2}$ state). We have also that

$$\frac{\alpha_s}{\alpha'_s} = (\Delta_s + 1)\frac{g_I}{g'_I}, \quad \text{and} \quad \frac{\alpha}{\alpha'} = \frac{g_I}{g'_I}.$$

Here we assume that the anomaly is the same for any s electron and zero for any non-s electron. This is not exactly true for $p_{1/2}$ electrons in general, but for the copper states of interest it is an excellent approximation. Using the above expressions we can obtain the results

TABLE V. The dipole hfs constant ratio in the different levels of the stable copper isotopes.

	$^2S_{1/2}$	$^4F_{9/2}$	$^4P_{5/2}$
$\frac{A^{63}}{A^{65}}$	0.933 568 5(0)[a]	0.933 551 6(16)	0.933 554 9(10)

[a]H. Figger, D. Schmidt, and S. Penselin (Ref. 5).

TABLE VI. The calculated value of $^{63}\Delta_s{}^{65}$ and $g_I{}^{63}/g_I{}^{65}$ for each combination of levels.

	$^2S_{1/2} - {}^4F_{9/2}$	$^2S_{1/2} - {}^4P_{5/2}$	$^4F_{9/2} - {}^4P_{5/2}$
$^{63}\Delta_s{}^{65}$	0.000 046(5)	0.000 050(5)	0.000 035(18)
$\frac{g_I{}^{63}}{g_I{}^{65}}$	0.933 526(5)	0.933 522(5)	0.933 532(20)

$$\frac{g_I}{g_I'}=\frac{(A/A')_1}{(\alpha_s/A)_1\Delta_s+1}=\frac{(A/A')_2}{(\alpha_s/A)_2\Delta_s+1},$$

$$\Delta_s=\frac{(A/A')_1-(A/A')_2}{(\alpha_s/A)_1(A/A')_2-(\alpha_s/A)_2(A/A')_1},$$

where 1 and 2 refer to different levels. α_s and α are obtained from the relations

$$A(^4F_{9/2})=\tfrac{1}{9}a_s+\tfrac{1}{3}a_p'+\tfrac{5}{9}a_d',$$

$$A(^4P_{5/2})=0.199a_s-0.473a_p'+0.840a_d'.$$

From the results in Table I we find

$$^{63}\Delta_s^{65}=0.000\,048(5),\quad \frac{g_I^{63}}{g_I^{65}}=0.933\,524(5).$$

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*Present Location: IBM Research Center, Yorktown Heights, New York.

†Present Location: New York University, University Heights, Bronx, New York.

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Correlation Properties of Light Scattered From Fluids*

L. Mandel

Department of Physics and Astronomy, The University of Rochester, Rochester, New York 14627

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The fluctuation and correlation properties of light scattered by a fluid are studied, without neglect of the spectral linewidth of the incident field. When the source is a single-mode laser, it is shown that, although the instantaneous amplitude of the scattered field obeys a Gaussian probability density, the scattered field is not a Gaussian field. The linewidth of the laser beam is reflected in the amplitude correlations of the scattered light, but not in the intensity correlations. On the other hand, when the laser is oscillating in more than one mode simultaneously, the spectral profile of the laser beam makes a contribution to the spectral density of the scattered intensity fluctuations, and cannot be neglected.

1. INTRODUCTION

The problem of light scattering by a fluid, particularly near the critical point, has been the subject of a great many experimental[1-14] and theoretical[14-25,4,8] investigations since the early work of Brillouin.[26] In recent years additional interest in the field was stimulated by the light beating technique developed by Benedek[3,4,6,7] and his coworkers, although light beating experiments were first reported by Forrester, Gudmundsen, and Johnson[27] in 1955. It has been