Title

COHERENCE MEASUREMENT OF TENSOR MULTIPOLES OF ASYMMETRICALLY EXCITED ATOMIC STATES

Authors

Church, D.A.
Michel, M.C.
Kolbe, W.

Publication Date

1975

Submitted to Physical Review Letters

LBL-3464 c.2
Preprint

COHERENCE MEASUREMENT OF TENSOR MULTIPOLES OF ASYMMETRICALLY EXCITED ATOMIC STATES

D. A. Church, M. C. Michel, and W. Kolbe

January 1975

Prepared for the U. S. Atomic Energy Commission
under Contract W-7405-ENG-48

COHERENCE MEASUREMENT OF TENSOR MULTIPOLES OF ASYMMETRICALLY EXCITED ATOMIC STATES

D.A. Church, M.C. Michel, and W. Kolbe
Lawrence Berkeley Laboratory, Berkeley, California 94720

Abstract

A specific coherence measurement technique is described, which yields the relative magnitudes of all predicted orientation and alignment parameters in tilted-foil-excited levels of ions and atoms. Quantum-beat frequencies and relative magnitudes are measured as a function of applied magnetic field strength and direction, and analyzed using Fano-Macek theory. The multipole moments of tilted-foil-excited 40 keV ^{4}He $4d\,^1D_2$ atoms are measured and related to proposed excitation mechanisms.

Recently we observed coherence properties of asymmetrically excited atoms and ions via quantum-beats in the optical decay radiation, when the uniformly moving excited particles were subject to an external uniform magnetic field oriented perpendicular to the beam direction.[1] Such beats also appear when the magnetic field is directed parallel to the ion beam.[2] We show here that measurements of the relative amplitudes of these quantum beats for both field directions, in linearly and circularly polarized light, completely define the relative magnitudes of the orientation and alignment tensors of the excited state. This technique is unambiguous, preserves the initial coherence, yields results with fractional per cent precisions, and applies to any excited level.

Equations describing the polarization and static angular distribution of electric dipole radiation in terms of orientation and alignment tensor parameters have been given by Fano and Macek.[3] In a uniform magnetic field, the predicted static radiation distribution is transformed to a time-dependent distribution by a coordinate rotation. Fano-Macek theory is based only on the symmetry properties of the collision geometry, rather than on any specific properties of the interaction. Nevertheless, the initial analysis of our quantum-beat data[1] showed effects not predicted by their equations. We have since learned[4] that one of the equations (18) of Ref. 3 is incorrect. An analysis of both original and new data, described below, is now in agreement with the corrected equations.

In terms of the orientation parameter O and the three alignment parameters A_o, A_1, and A_2 defined for the detection (det) and collision (col) reference frames, the corrected equations (18) from Ref. 3 are:

$$O_o^{det} = O_{1-}^{col} \sin\theta \sin\phi$$

$$A_o^{det} = \frac{1}{2} A_o^{col} (3\cos^2\theta - 1) + \frac{3}{2} A_{1+}^{col} \sin 2\theta \cos\phi + \frac{3}{2} A_{2+}^{col} \sin^2\theta \cos 2\phi$$

$$A_{2+}^{det} = \frac{1}{2} A_o^{col} \sin^2\theta \cos 2\psi + A_{1+}^{col} \{\sin\theta \sin\phi \sin 2\psi + \sin\theta \cos\theta \cos\phi \cos 2\psi\}$$

$$+ A_{2+}^{col} \{\tfrac{1}{2}(1+\cos^2\theta) \cos 2\phi \cos 2\psi - \cos\theta \sin 2\phi \sin 2\psi\}$$

where θ, ϕ are the polar coordinates of the light detector in a coordinate system which has as its $\hat{z}$ axis the ion beam axis (see e.g. Fig. 1 of Ref. 1). The orientation angle of a linear polarization analyzer relative to the $\hat{z}$ axis is ψ. In our geometry, $\theta = \phi = \pi/2$. If an external uniform magnetic field is applied, the multipole moments of the excited states precess in time t about the field direction at the Larmor frequency ω. For a field parallel to the beam direction $\hat{z}$, $\theta = \pi/2$ and $\phi = \pi/2 + \omega t$, while a field perpendicular both to the beam and the direction of light observation $\hat{y}$ produces

$\theta = \pi/2 + \omega t$ and $\phi = \pi/2$. The expression for the optical decay light intensity (Eqn. 14, Ref. 3) for circularly polarized light in a given transition is

$$I_{cp} = \frac{1}{3} CS\{1 - \frac{1}{2} h^{(2)} (j_i,j_f) A_o^{det} + \frac{3}{2} h^{(1)} (j_i,j_f) O_o^{det} \tag{1}$$

while the corresponding expression for linearly polarized light is

$$I_{\ell p} = \frac{1}{3} CS\{1 - \frac{1}{2} h^{(2)} (j_i,j_f) A_o^{det} + \frac{3}{2} h^{(2)} (j_i,j_f) A_{2+}^{det} \tag{2}$$

The $h^{(k)}(j_i,j_f)$ are ratios of 6j-coefficients and the C and S are constants for a given geometry and transition[3]. For the 2p $^1P_1$- 4d $^1D_2$ transition of ^{4}He I, $h^{(1)}(2,1) = +3$ and $h^{(2)}(2,1) = -1$. Substituting the expressions for the multipole parameters and angles into equations (1) and (2) above, for each of the two magnetic field directions, we obtain for magnetic field parallel to the beam ($\equiv H\|$)

$$I_{cp} = \frac{1}{3} CS\{1 - \frac{1}{4} A_o^{col} - \frac{3}{4} A_{2+}^{col} \cos 2\omega t + \frac{9}{2} O_{1-}^{col} \cos\omega t\} \tag{3}$$

$$\begin{aligned} I_{\ell p} = \frac{1}{3} CS\{1 &- \frac{1}{4} A_o^{col} (1+3\cos 2\psi) - \frac{3}{2} A_{1+}^{col} \cos\omega t \sin 2\psi \\ &- \frac{3}{4} A_{2+}^{col} \cos 2\omega t (1-\cos 2\psi)\} \end{aligned} \tag{4}$$

and for H perpendicular to the beam ($\equiv H\perp$)

$$I_{cp} = \frac{1}{3} CS\{1 + \frac{1}{8} (A_o^{col} - 3A_{2+}^{col}) - \frac{3}{8} (A_o^{col} + A_{2+}^{col}) \cos 2\omega t + \frac{9}{2} O_{1-}^{col} \cos\omega t\} \tag{5}$$

$$\begin{aligned} I_{\ell p} = \frac{1}{3} CS\{1 &+ \frac{1}{8} (A_o^{col} - 3A_{2+}^{col})(1-3\cos 2\psi) - \frac{3}{2} A_{1+}^{col} \cos\omega t \sin 2\psi \\ &- \frac{3}{8}(A_o^{col} + A_{2+}^{col}) \cos 2\omega t (1+\cos 2\psi)\} \end{aligned} \tag{6}$$

Each orientation and alignment parameter, or in certain instances a sum of parameters, is distinguished by unique frequency and polarization dependences of the decay radiation intensity. In particular, each component varying at ω

in linearly polarized light arises from the alignment component A_{1+}^{col} rather than from an orientation coherence, as originally hypothesized[1]. Alignment components varying at 2ω have a $(1 - \cos 2\psi)$ dependence when H is parallel to the beam, and a $(1 + \cos 2\psi)$ dependence when H is perpendicular to the beam. These results are quite general.

The mean values of each of the four total absolute intensities (Eqns. 3-6) depend on the relative magnitudes of the observed orientation and alignment parameters, although the sinusoidally varying terms do average to zero over an integral number of periods. In practical beam-tilted-foil experiments, the relative magnitudes of the multipole parameters are generally $\lesssim 0.1$, so the normalized <u>relative</u> amplitude of each frequency component can be determined to $\approx 10^{-2}$ even without correction by an iteration procedure. This uncertainty is comparable to usual statistical error.

Using equations 3-6 we have analyzed the orientation and alignment of the 4d $^1D_2$ level of ^{4}He I, observed via the 4922 Å transition, produced by passing 40 keV ^{4}He ions through thin ($\approx 6 \mu g/cm^2$) carbon foils with exit surface normal at an angle $\beta = 30$ degrees from the beam direction (see Fig. 1 of Ref. 1). Light emitted from the atoms a fixed distance d downstream from the foil was collected from a narrow spatial region parallel to the foil surface. The time relative to excitation $t = d/v$ is fixed by the mean atom velocity v. The collected light was polarization analyzed, spectrum analyzed, and measured as a function of the linearly swept magnetic field strength H. The periodically varying intensity was fitted to a 5-parameter (3 amplitudes, 2 frequencies) curve with provision also taken for amplitude attenuation produced by any dephasing. Relatively insignificant corrections to the raw data, of the order of a few per cent of total light intensities, were ignored. Such corrections arise from a low constant background, weak incoherent cascading to the upper level, and imperfections in the polarization analyzers.

The data are plotted in Fig. 1 as a function of linear polarization analyzer angle ψ, together with results of measurements with a circular polarizer. Theoretically predicted angular dependences, normalized to the data, are indicated by solid or broken curves. The uncertainty of an individual datum is about ± 0.5%. The analyzed data of Fig. 1 are also tabulated in Table 1-a. The orientation and alignment parameters are in some cases over-determined by the data, permitting checks for consistancy. One sees the agreement between different measurements of the same quantity. The parameter A_o^{col} is determined only by subtracting the independently measured value of A_{2+}^{col}, and consequently has the largest uncertainty.

Variations of the alignment and orientation parameter magnitudes with foil tilt angle will be predicted by specific interaction theories. Table I-b shows the mean magnitudes of the parameters 0_{1-}^{col}, A_{1+}^{col} and A_{2+}^{col} at foil angles β of 30, 45 and 60 degrees. All three of these components increase monotonically with β. A_o^{col} decreases with increasing β, but our current data are inadequate to establish a dependence. At $\beta = \pi/4$, A_o^{col} has decreased to about 80% of its value at $\beta = 0$.

We have attempted to fit the data of Table I-b to different powers of the elementary trigonometric functions. It seems plausible to rule out those functions, such as tan β, that increase without limit near $\beta = \pi/2$, since the relative values of the moments must remain ≤ 1. With this restriction, we find that 0_{1-}^{col} is proportional to $\sin^2\beta$ (see Fig. 2), and $A_{1+}^{col} \propto \sin\beta$. These different functional dependencies again clearly distinguish orientation and alignment parameters which vary at the same frequency. Our fit gives

$A^{col}_{2+} \propto \sin^3\beta$, but this high power should be tested with additional data at large angles. That the orientation vector is odd[1] in β, does not preclude the $\sin^2\beta$ behavior illustrated in Fig. 2; the amplitude of the beat is independent of its phase.

Some of these tilt-angle dependences can be directly related to theoretical predictions. On the basis of a simple torque model[5], one might expect an angular dependence of the orientation proportional to $\sin\beta$. The possibility that the coherent orientation is generated by an electric field normal to the foil surface has been proposed[1,6] and analyzed for a $^1P_1$ level by Eck[6]. This analysis is presented in terms of relative sub-level cross-sections and related to the Stokes parameters[7], which are not in general simply related to the multipole parameters[8]. However, the orientation is directly proportional to the Stokes parameter S. Eck predicts a complex variation of orientation with β for a $^1D_2$ level[8], but the expression is overall multiplied by $\sin 2\beta$ in contrast to the results in Fig. 2.

The measurement and analysis of quantum-beat data in terms of multipole parameters by our technique is directly applicable in the low and high field limits to levels with hyperfine or other interactions. In certain instances it is found that the orientation of the level is partially destroyed by motional electric fields when H is perpendicular to the beam[2]. Since the magnitude of such an effect is a function of the time relative to excitation, extrapolation of the data to a standard value permits analysis by the method presented here.

References

1. D.A. Church, W. Kolbe, M.C. Michel, and T. Hadeishi, Phys. Rev. Letters <u>33</u>, 565 (1974).

2. C.H. Liu, S. Bashkin, and D.A. Church, Phys. Rev. Letters 33, 993 (1974).

3. U. Fano and J.H. Macek, Rev. Mod. Phys. 45, 553 (1973).

4. J. Macek (private communication).

5. S. Bashkin (private communication).

6. T.G. Eck, Phys. Rev. Letters 33, 1055 (1974).

7. H.G. Berry, L.J. Curtis, D.G. Ellis, and R.M. Schectman, Phys. Rev. Letters 32, 751 (1974).

8. T.G. Eck (private communication).

Figure Captions

Fig. 1. Relative amplitudes of ω and 2ω beats plotted as a function of the angle of the linear polarization analyzer ψ relative to the beam ($\hat{z}$) axis. Circular polarization data are also shown by interrupted horizontal points. The solid and dashed curves are theoretical predictions from equations 3-6 normalized to the data. The foil tilt angle $\beta = 30$ degrees.

Fig. 2. The relative amplitude of the ω beats in circularly polarized light, proportional to the orientation, plotted vs. $\sin^2\beta$, where β is the foil tilt angle.

TABLE I

(a) Spherical tensor moments of the 4d $^1D_2$ level of He I following excitation of a 40 keV beam by a carbon foil with normal at angle β = 30 degrees to the beam direction. Components obtained with particular polarization analyzers and magnetic field directions are distinguished. Equations 3-6 are used in the analysis.

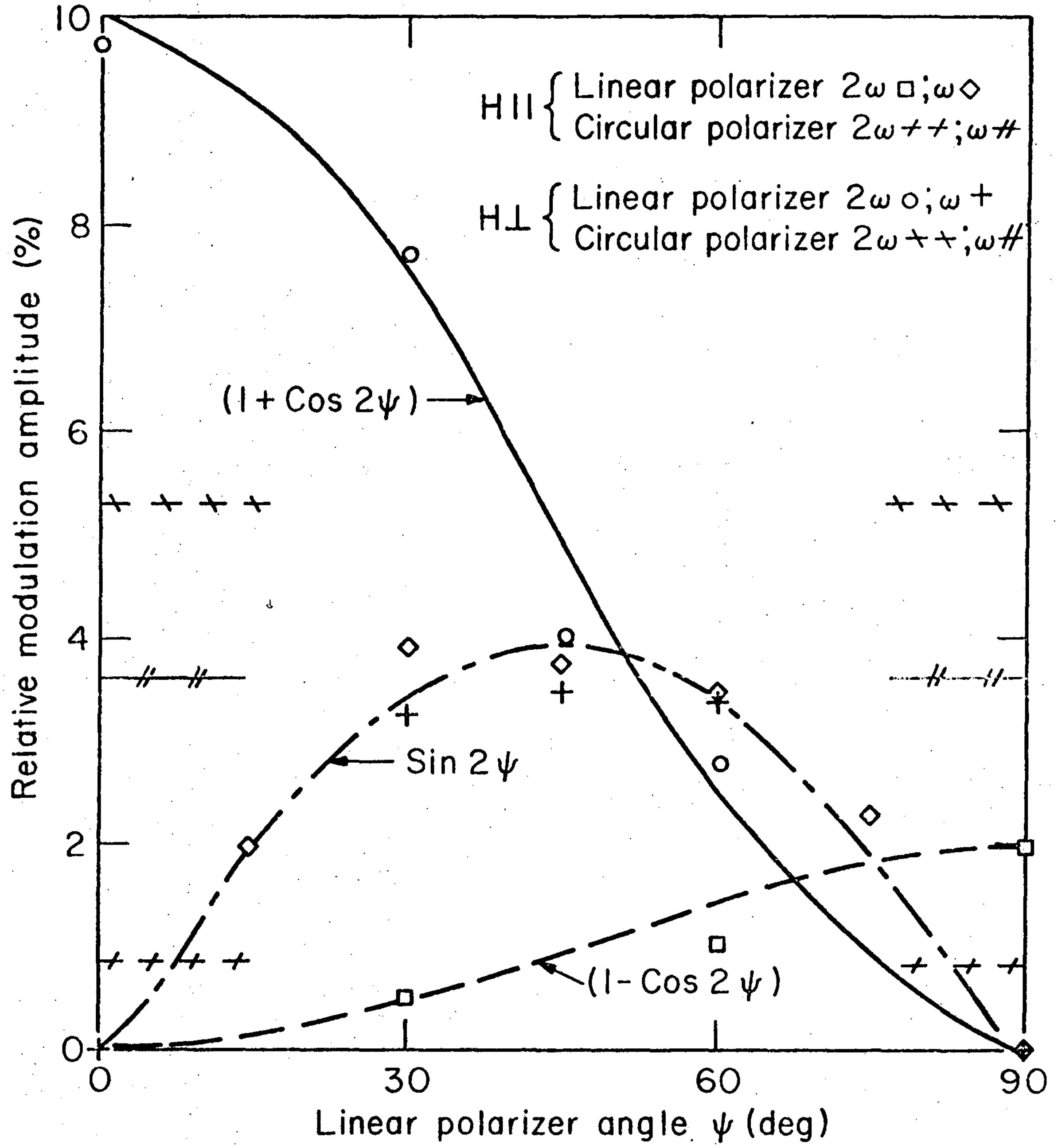
	Linear Polarizer		Circular Polarizer	
H∥	A^{col}_{2+}	-0.013	A^{col}_{2+}	-0.011
	A^{col}_{1+}	-0.027	O^{col}_{1-}	-0.008
H⊥	$A^{col}_{2+}+A^{col}_{o}$	-0.133	$A^{col}_{2+}+A^{col}_{o}$	-0.141
	A^{col}_{1+}	-0.027	O^{col}_{1-}	-0.008
	A^{col}_{o}	-0.12[a]	A^{col}_{o}	-0.13[a]

[a] value derived from the sum of A^{col}_{2+} + A^{col}_{o}.

(b) Relative tensor components derived from equations 3-6 at different foil tilt angles β.

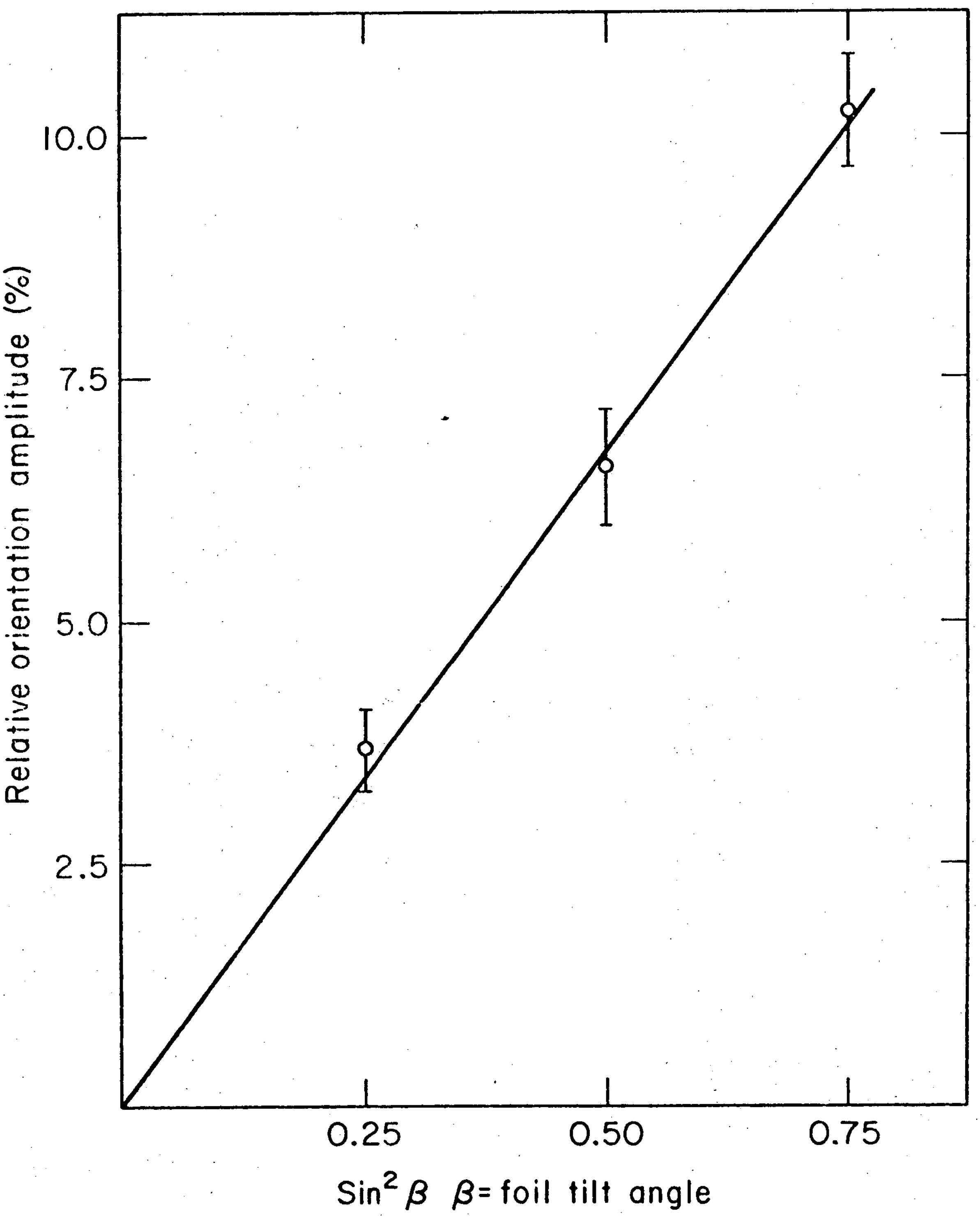
		β (degrees)		
		30	45	60
Tensor component	O^{col}_{1-}	-0.008	-0.015	-0.023
	A^{col}_{1+}	-0.027	-0.037	-0.05
	A^{col}_{2+}	-0.012[b]	-0.025[b]	-0.053[b]

[b] mean of two separate measurements.



XBL 751-2020

Fig. 1



XBL751-2019

Fig. 2

TECHNICAL INFORMATION DIVISION
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720