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Quantum interference in two-photon absorption: Polarization and magnetic field effects in the $(7s)^1S \leftarrow (5s^2)^1S$ transition of atomic Sr

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Quantum-mechanical interference is found in the two-photon excitation of an atomic ^1S state through the nonresonant sequence $^1S \leftarrow {}^1P \leftarrow {}^1S$, since the magnetic sublevels of the intermediate ^1P state provide multiple pathways for absorption. Depending on the relative polarization of the two incident light beams, the interference can be either constructive or destructive. When the electric vectors of the light beams are linearly polarized and orthogonal, the interference is purely destructive and two-photon transitions are forbidden. Application of an external magnetic field, however, lifts the degeneracy of the intermediate state giving a two-photon absorption rate proportional to the square of the magnetic field strength. These effects are reported for the $(7s)^1S \leftarrow (5s^2)^1S$ transition of atomic Sr excited by two dye lasers operating at different wavelengths.

It is well known in quantum theory that the existence of two paths leading to an event gives rise to interference in the probability for the outcome of the event, provided the two pathways cannot be distinguished in an experiment. In the case of two-photon absorption where one light beam with frequency Ω_1 is nearly coincident with a ^1P-^1S energy splitting [see Fig. 1(a)] and a second beam with frequency Ω_2 is tuned such that $\Omega_1+\Omega_2$ coincides with a ^1S-^1S splitting, provided the polarization vectors of the light beams are orthogonal, then the interference between the two pathways for absorption of radiation is destructive so that two-photon transitions are forbidden. If a magnetic field is applied in a direction perpendicular to the plane formed by the two electric vectors of the light beams, the degeneracy of the intermediate ^1P state is lifted and the interference becomes partially constructive, leading to a finite two-photon transition probability. A detailed description of the effect of the relative orientation of the electric vectors of the two light beams as well as the dependence of the transition probability on the magnetic field strength has been given in Ref. 1. Here, observation of the interference effect is reported in the two-photon excitation of the $(7s)^1S$ state of atomic Sr from the $(5s^2)^1S$ ground state using two dye-laser beams.

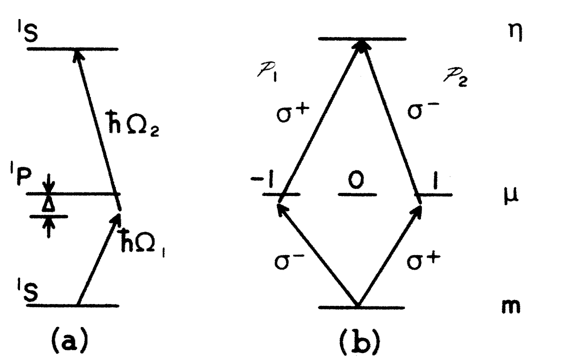

FIG. 1. (a) Energy-level diagram for two-photon absorption. The photon energy of the first beam $\hbar\Omega_1$ is detuned an amount Δ from a ^1P level. (b) The same energy-level diagram as in (a) showing the two pathways $\mathcal{P}_1$ and $\mathcal{P}_2$ for two-photon population of the upper ^1S state.

Consider an atom in a magnetic field (which defines the quantization axis Z) that is irradiated with light linearly polarized perpendicular to Z and whose frequency is detuned from an intermediate state transition by an amount Δ. The effect of such σ radiation is to excite a time-varying polarization in the intermediate state that is a coherent superposition of the $m_1=+1$ and $m_1=-1$ sublevels of the intermediate state. If each of the linearly polarized radiation beams is expressed in terms of superpositions of left- and right-hand circular polarizations, then there are two paths denoted as $\mathcal{P}_1$ and $\mathcal{P}_2$ in Fig. 1(b) for excitation of the upper state η. In the path $\mathcal{P}_1$, the upper state is connected to the ground state through the σ^- component of the first radiation beam, and the σ^+ component of the second beam; in $\mathcal{P}_2$ the reverse is true. Both paths have identical probability amplitudes but different phases; thus, when the transition probability between states m and η is computed, interference between the two pathways is found. For an arbitrary orientation of the electric vectors of the two light beams, the transition probability between the ground and excited states is given by second-order perturbation theory as[1]

$$R=\pi\sum_{\mu,\mu'}\frac{F_{\mu\mu'}G_{\mu'\mu}\delta(\omega_{\eta m}-(\Omega_1+\Omega_2))}{[(\omega_\mu-\Omega_1)-i\Gamma/2][(\omega_{\mu'}-\Omega_1)+i\Gamma/2]}\ , \qquad (1)$$

where δ is the Dirac delta function, ω_μ and $\omega_{\eta\mu}$ are the frequencies corresponding to the intermediate–ground-state-energy splitting (as perturbed by the external field) and the upper-state–ground-state splittings, respectively, and Γ is the radiative decay constant for the intermediate state. The matrices F and G are defined by

$$F_{\mu\mu'}=H^f_{\mu m}H^f_{m\mu'},\ G_{\mu'\mu}=H^g_{\mu\eta}H^g_{\eta\mu'}\ ,$$

where $H^f_{\mu m}$ and $H^g_{\mu\eta}$ are electric dipole matrix elements for the first and second light beams with polarizations $\hat{\mathbf{f}}$ and $\hat{\mathbf{g}}$, respectively. In Eq. (1) the dependence of the transition rate on the orientation of the electric field vectors is contained in the matrices F and G. The effect of external fields is to change the atomic energy-level spacings which affects the values of ω_μ in the denominator of Eq. (1). For the case of two-photon transitions between ^1S states with an intermediate ^1P state, Eq. (1) reduces to

$$R = I_1 I_2 \frac{(g\beta H/\hbar)^2\cos^2\phi_g + (\Delta^2+\Gamma^2/4)\sin^2\phi_g + (g\beta H\Gamma/\hbar)\cos\phi_g\sin\phi_g}{[(\Delta+g\beta H/\hbar)^2+\Gamma^2/4][(\Delta-g\beta H/\hbar)^2+\Gamma^2/4]} , \quad (2)$$

where $\phi_g+\pi/2$ is the polar angle between the polarization vectors of the two light beams in the XY plane, β is the Bohr magneton, $\hbar$ is Planck's constant divided by 2π, g is the Landé factor of the intermediate state, H is the magnetic field strength, and I_1 and I_2 are the intensities of the two light beams.

From Eq. (2) it is clear that with two orthogonal electric vectors ($\phi_g=0$) the interference is destructive and R is zero; in addition, for large values of Δ (i.e., $\Delta \gg g\beta H/\hbar$), R increases as the square of the magnetic field strength. On the other hand, for parallel electric vectors ($\phi_g=\pi/2$) the interference is constructive and R is proportional to $\Delta^2+(\Gamma/2)^2$. Thus, at a fixed magnetic field strength, continuous rotation of the relative electric field polarizations gives alternately constructive then partially destructive interference which can be monitored by measuring the population of the excited ^1S state.

Experiments were carried out by exciting an atomic beam of Sr with the output of two linearly polarized dye lasers pumped by a single N_2 laser (see Fig. 2). The first dye laser (Molectron Inc., DL14) was operated at 460.7 nm and was tuned to be nearly coincident with the $(5p)\,^1P \leftarrow (5s^2)\,^1S$ transition. This laser was operated with a pressure tuned intracavity etalon and had a bandwidth (full width at half maximum) of 1.0 GHz as measured by a scanning Fabry-Perot interferometer. The second dye laser was tuned to 597.2 nm, which along with the first dye laser excited Sr to the $(7s)\,^1S$ state. The second laser, based on a double-grating grazing incidence design,[2,3] was used without additional bandwidth narrowing optics. Both dye laser beams were passed through linear polarizers and combined with a high reflectivity dichroic mirror. A quartz half-wave plate was placed in the path of the 461-nm beam immediately preceding the dichroic mirror. The half-wave plate was then rotated about its axis by a stepping motor so that the relative polarization directions of the two beams could be varied.

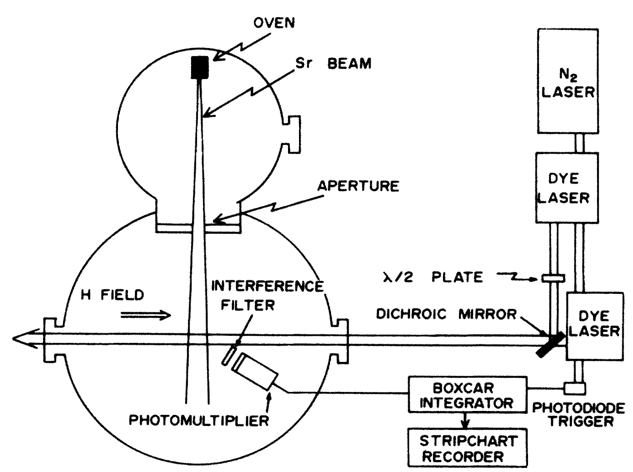

FIG. 2. Diagram of the experimental apparatus. Fixed magnetic fields were produced by varying the gap in a permanent magnet placed in the vacuum chamber.

The colinearly propagating laser beams were directed into a differentially pumped high vacuum chamber where they crossed an atomic Sr beam at perpendicular incidence. The atomic beam (produced by vaporization of metallic Sr in a resistively heated oven at 900 K) was collimated by a pinhole between the two vacuum chambers so that its beam divergence corresponded to a Doppler width approximately equal to the natural linewidth (20 MHz) of the ^1P-^1S transition. The population of the upper ^1S state was measured by recording the fluorescence intensity of the $(7s)\,^1S \rightarrow (5p)\,^3P$ transition[4] at 414 nm. A photomultiplier (EMI Inc., Model 9829 QB) viewed the region of intersection of the laser beams with the atomic beam in the center of the magnetic field through a narrow-band interference filter; in addition, a 1-cm aperture was employed to restrict the field of view of the photomultiplier, thus reducing the signal from scattered laser light. Although the transition probability for the 414-nm line is comparatively small, adequate signal-to-noise ratios could be obtained in the photomultiplier output since a high proportion of the scattered laser light was rejected. The signal from the photomultipler was fed to a boxcar averager (EG&G-PAR Inc., Model 162-164) whose output was displayed on a strip-chart recorder.

Data were taken by recording the output of the boxcar averager as the half-wave plate was slowly rotated for fixed values of the magnetic field. Figure 3 shows typical signals obtained at a detuning Δ corresponding to 3.4 GHz with the field on (a) and with the field off (b). According to Eq. (2), the ratio of the signal with the beams perpendicular $R_\perp$ to the signal with the beams parallel, $R_\parallel$ is given by

$$R_\perp/R_\parallel = (g\beta H/\hbar)^2/(\Delta^2+\Gamma^2/4) . \quad (3)$$

By recording data as shown in Fig. 3 for a fixed value of Δ at several different field settings, a plot of $R_\perp/R_\parallel$ versus the magnetic field strength was generated. Figure 4 is such a plot for two different values of the detuning showing the quadratic dependence of $R_\perp/R_\parallel$ with the magnetic field. Although measurement of a Landé factor was not the purpose of these experiments, a least-squares[5] fit to the data in Fig. 4 gave a value[6] of 1.01 ± 0.06 for the $(5p)\,^1P$ state. The most important source of error[7] in the determination of this figure comes from the uncertainty in the experimental value of Δ. Note that for the large values of the detuning used here, Δ^2 is much greater than $\Gamma^2/4$, so that the Landé factor measurement is referenced only to the detuning.

At high laser intensities, the ac Stark effect shifts the atomic energy levels from their zero field values. Since Eqs. (1) and (2) were derived under the assumption that the detuning is dependent on the external magnetic field

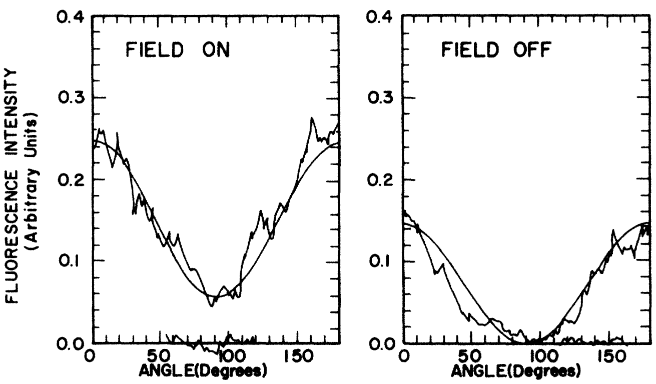

FIG. 3. Fluorescence intensity (in arbitrary units) as a function of the angle between the electric vectors of the two dye lasers for a detuning of 3.4 GHz. In (a), a magnetic field of 0.127 T has been applied; in (b) the field is absent. The abscissa is the angle between the polarization vectors of the two laser beams: at 0° and 180° the electric vectors are parallel (i.e., $\phi_g = -\pi/2$ and $\pi/2$, respectively); at 90° the two electric vectors are perpendicular ($\phi_g = 0$). The baselines are recorded with the 461-nm laser beam tuned off of the atomic resonance. The solid curves are plotted from Eq. (2).

alone, it is appropriate to estimate the magnitude of the Stark shift produced by the laser beam at 460.7 nm. This can be done using the result from first-order perturbation theory for the quadratic Stark effect,[8] with the value for the electric dipole matrix element obtained from the f number for the transition.[9,10] The relatively low power density in the dye laser beam of approximately 5×10^5 W/m^2 corresponds to a shift of 200 MHz (for a detuning of 5 GHz), which is less than the error in the determination of Δ.

The two-photon interference effect[11] described here is in many ways analogous to the Hanle effect[12-14] which is frequently used for the measurement of Landé factors.

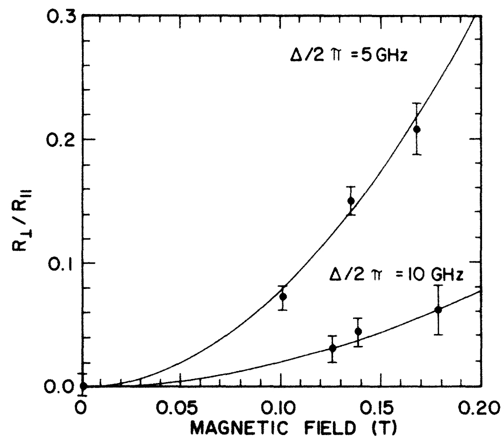

FIG. 4. Plot of $R_\perp/R_\parallel$ vs magnetic field for two values of the detuning. The magnetic field strength was measured with a calibrated Hall probe. The error bars represent statistical errors for several measurements at each field point. The solid lines are calculated from the least-squares fit to the data using Eq. (2).

Indeed, Eq. (1) bears a strong resemblance to the Breit-Franken equation[14] that gives the quantum mechanical description of the Hanle effect. Both phenomena arise from the production of a coherent superposition of the intermediate states ($\mu = \pm 1$ in the present case) so that two indistinguishable pathways to the final state exist. In both cases, the interference is constructive or destructive depending on the relative phases for the two pathways, which are determined by the polarization and propagation directions for the two light beams (i.e., the incident and scattered light beams in the case of the Hanle effect).

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[6]We have been unable to find reference to a previous measurement of Landé factor for this state. Note that A. Lurio, R. De Zafra, and R. Goshen, Phys. Rev. **134**, A1198 (1964), have assumed the Landé factor for ^1P Sr to be unity.

[7]With the two dye lasers used here, the isotopic shifts in the different isotopes of Sr (relative to ^{88}Sr) could not be resolved. However, for fields in excess of 0.1 T, the effect of naturally occurring ^{87}Sr (7% abundance) with a nuclear spin of $\frac{9}{2}$ should be of no consequence since the magnetic field splittings in the Back-Goudsmit limit far exceed the hyperfine splittings (46 MHz) of this isotope. Although the zero field splitting of ^{84}Sr is greater than this, its natural abundance is sufficiently small (0.6%) so that it should give an insignificant signal compared with that from ^{88}Sr. See D. Bender, H. Brand, and V. Pfeiffer, Z. Phys. A **318**, 219 (1984); or T. Whitaker (private communication) for the values of the isotopic splitting of Sr. Hyperfine splitting constants can be found in A. J. Kluge and H. Sauter, Z. Phys. **270**, 295 (1974).

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