

electric dipole transition slightly allowed as well. The transition amplitude for this interaction is of the form

$$A_{PNC}(F, m; F', m') = e^{i\phi^{\omega_1}} \{ \varepsilon_z^{\omega_1} \delta_{m, m'} + [\pm \varepsilon_x^{\omega_1} + i \varepsilon_y^{\omega_1}] \delta_{m, m' \pm 1} \} i \text{Im}(\mathcal{E}_{PNC}) C_{F, m}^{F', m'}, \quad (3)$$

where \mathcal{E}_{PNC} is the matrix element for electric dipole transitions due to the state mixing by the PNC interactions. To measure A_{M1} or A_{PNC} directly is problematic in that their magnitudes are typically well below the level of measurement noise. Techniques using an interference between the weak transition and a stronger transition (the Stark-induced transition, for example) have therefore been developed to effectively amplify the signal to a detectable level. For example, under conditions that allow a strong Stark-induced amplitude and a weak PNC amplitude on the same transition that add constructively, the net rate scales as $W_+ = |A_{St} + A_{PNC}|^2 \simeq |A_{St}|^2 + 2|A_{St}||A_{PNC}|$. The interference between these various amplitudes can be reversed by reversing one or more of the fields that influences the sign of the amplitudes, resulting in a rate $W_- = |A_{St} - A_{PNC}|^2 \simeq |A_{St}|^2 - 2|A_{St}||A_{PNC}|$. A precise measurement of the small difference between W_+ and W_- can then be used to determine $|A_{PNC}|$.

In each of the previous measurements, a single laser field has been employed, and both interactions (strong and weak) are linear in the amplitude of this field. We now consider this system under the influence of a second optical field composed of components at frequencies ω_2 and ω_3 , where $\omega_2 + \omega_3 = \omega_1$, which is capable of driving the $6s \rightarrow n's$ transition via a two-photon interaction. In order for these amplitudes to interfere, the ω_1 field component must be phase coherent with ω_2 and ω_3 components, as it will be when the former is generated from the latter using a nonlinear optical crystal for sum frequency generation. We have previously demonstrated this interference between two-photon absorption and Stark-induced linear absorption on the cesium $6s \rightarrow 8s$ transition [26]. We write the transition amplitude for this interaction in a form similar to that of the Stark-induced transition given by Eq. (1),

$$A_{2PA}(F, m; F', m') = e^{i(\phi^{\omega_2} + \phi^{\omega_3})} \times \left\{ \left[\tilde{\alpha} \varepsilon^{\omega_2} \cdot \varepsilon^{\omega_3} \delta_{F, F'} + i \tilde{\beta} (\varepsilon^{\omega_2} \times \varepsilon^{\omega_3})_z C_{F, m}^{F', m'} \right] \delta_{m, m'} + \tilde{\beta} \left[\pm i (\varepsilon^{\omega_2} \times \varepsilon^{\omega_3})_x - (\varepsilon^{\omega_2} \times \varepsilon^{\omega_3})_y \right] C_{F, m}^{F', m'} \delta_{m, m' \pm 1} \right\}, \quad (4)$$

where ε^{ω_2} and ε^{ω_3} are the amplitudes, and phases ϕ^{ω_2} and ϕ^{ω_3} , the phases, of the optical waves at frequencies ω_2 and ω_3 , respectively, and the coefficients of the two-photon moments, $\tilde{\alpha}$ and $\tilde{\beta}$, are defined in a form similar to the Stark polarizabilities. Bouchiat and Bouchiat [25] noted the relationship between Stark-induced transitions and two-photon absorption. The interference between the two-photon amplitude and the amplitudes that are linear in ε^{ω_1} can be observed on $\Delta F = 0$ as

well as $\Delta F = \pm 1$ transitions. The $\Delta F = 0$ transitions, however, present the following two advantages over $\Delta F = \pm 1$ transitions: (1) Systematic errors due to magnetic dipole contributions to \mathcal{E}_{PNC} are smaller, and (2) the two frequencies ω_3 and ω_2 can be equal, so this measurement requires only a single laser source and the ω_1 beam can be generated by frequency doubling the ω_2 laser output in a nonlinear crystal. We will consider only $\Delta F = 0$, $\Delta m = 0$ transitions in the following, and write the two-photon transition amplitude of Eq. (4) as $A_{2PA} = \tilde{\alpha} (\varepsilon^{\omega_2})^2 e^{2i\phi^{\omega_2}}$.

To maintain a constant phase difference between the two-photon amplitude and the linear amplitudes, the optical beams must propagate in directions nearly co-linear with one another. Without loss of generality, we define the y -axis along \mathbf{k} , such that $\varepsilon_y^{\omega_1}$ must vanish for a plane wave or weakly-focussed beam. We allow an arbitrary static electric field $\mathbf{E} = E_x \hat{x} + E_y \hat{y} + E_z \hat{z}$, and consider a dc magnetic field \mathbf{B} that is primarily in the \hat{z} -direction. \mathbf{B} will separate the various m -components: $\Delta E_{F, m} = \mu_B g_F m B_z$, where μ_B is the Bohr magneton and g_F is 1/4 for $F=4$ and -1/4 for $F=3$. The effect of the \hat{x} - and \hat{y} -components for \mathbf{B} , which can be present in an experiment due to imperfect alignment, is to mix the magnetic sublevels

$$\begin{aligned} |ns^2 S_{1/2}, F, m\rangle &= |ns^2 S_{1/2}, F, m\rangle \\ &+ |ns^2 S_{1/2}, F, m-1\rangle \frac{B_x + i B_y}{B_z} C_{F, m}^{F, m-1} \\ &- |ns^2 S_{1/2}, F, m+1\rangle \frac{B_x - i B_y}{B_z} C_{F, m}^{F, m+1}. \end{aligned} \quad (5)$$

We include in this expression mixing among magnetic components of the same F , but omit mixing with other F states, an approximation that will be valid for the modest magnetic field strengths characteristic of these measurements.

We sum the four transition amplitudes,

$$\begin{aligned} \sum A &= A_{2PA} + \left\{ \left[\alpha E_z \varepsilon_z^{\omega_1} + \alpha E_x \varepsilon_x^{\omega_1} - M \varepsilon_x^{\omega_1} C_{F, m}^{F, m} \right. \right. \\ &\quad \left. \left. - M \varepsilon_z^{\omega_1} \frac{B_x}{B_z} \Delta C^{(2)} \right] + i \left\{ \text{Im}(\mathcal{E}_{PNC}) \varepsilon_z^{\omega_1} C_{F, m}^{F, m} \right. \right. \\ &\quad \left. \left. - \beta E_y \varepsilon_x^{\omega_1} C_{F, m}^{F, m} + \beta E_y \varepsilon_z^{\omega_1} \frac{B_x}{B_z} \Delta C^{(2)} \right. \right. \\ &\quad \left. \left. + \beta E_z \varepsilon_x^{\omega_1} \frac{B_y}{B_z} \Delta C^{(2)} - \beta E_x \varepsilon_z^{\omega_1} \frac{B_y}{B_z} \Delta C^{(2)} \right\} \right] e^{i\phi^{\omega_1}}, \end{aligned} \quad (6)$$

where all terms except $\varepsilon_x^{\omega_1}$ are real, and $\Delta C^{(2)} = \sum_{+/-} \left\{ (C_{F, m}^{F, m \pm 1})^2 - C_{F, m \pm 1}^{F, m} C_{F, m}^{F, m \pm 1} \right\}$ is 3/16 for $F = 3$, $m = \pm 3$, and 1/4 for $F = 4$, $m = \pm 4$. This factor is small, but not negligible, in comparison to $C_{F, m}^{F, m} = \mp 3/4$ for $F = 3$, $m = \pm 3$ or ± 1 for $F = 4$, $m = \pm 4$. The relative scale of the different amplitudes in Eq. (6) depends on many factors, but for $E_y < 100$ V/cm, the two-photon rate dominates all others, even with cw beam powers