tremely simple technique to use, requiring a bare minimum of optical components.

The possibility of performing Doppler-free intermodulated spectroscopy in an electrodeless radio-frequency discharge deserves careful investigation. Superregenerative radio-frequency detection may provide a useful form of optogalvanic detection.

In conclusion, intermodulated optogalvanic spectroscopy has been demonstrated to be a powerful new method of performing Doppler-free saturation spectroscopy in a discharge. We report preliminary hyperfine splittings of the 3^3D level of ^{3}He obtained using this method. Finally, our calculations indicate that IMOGS compares favorably in sensitivity to other Doppler-free techniques.

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Extended Two-Level Theory of the Exponential Index of Multiphoton Processes

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A nonperturbative formula is given for the r-photon-resonant, $(r+p)$-photon ionization rate of an atom and from it is derived an expression for the exponential index.

The exponential order, or index, of an atomic multiphoton process is defined by

$$K = \partial \log(\text{rate})/\partial \log(\text{intensity}). \quad (1)$$

It provides a convenient way to parametrize experimental data[1] because it does not require an absolute measurement of either the observed rate or the inducing laser's intensity. Clearly if the rate is simply proportional to the intensity, $R = \text{const} \times I^n$, then one has $K = n$, for all I. However, it is found experimentally that not only R but K also depends on I. This occurs when the laser frequency is such that an integer number of laser photons and an allowed atomic transition are near to resonance.

In spite of the phenomenological utility of the index K, there is no satisfactory theory for it. A satisfactory theory should be able to (a) apply both to ionization and to purely bound-bound multiphoton processes; (b) express K analytically and in terms of parameters significantly simpler than K itself; (c) treat the region very near to resonance; (d) apply at laser intensities both above and below the saturation level; and (e) accommodate standard line-broadening mechanisms (spontaneous emission, collisions, laser bandwidth, Doppler effect). The purpose of this note is to present a theory for multiphoton rates that

meets all five criteria, and from which the exponent K can easily be obtained. The theory is based on an extended two-level (ETL) rate model for high-intensity near-resonant absorption.

I will use, as far as possible, the notation of the basic nonperturbative two-level rate model previously discussed.[2] The ETL model atom is shown in Fig. 1. The quantity of experimental interest is the rate[3] of transfer of population from the ground state $|1\rangle$ to the final broad "continuum" state $|c\rangle$ via an intermediate r-photon-resonant state $|2\rangle$. Coherent Rabi oscillations may or may not occur in the r-photon transition, but are assumed absent in the p-photon transition because of the breadth of $|c\rangle$.

The central constituents of the ETL rate are the r-photon saturable bound-bound rate $R_{12}^{(r)}$, and the p-photon rate $R_{2c}^{(p)}$ from $|2\rangle$ to $|c\rangle$. These rates are taken to be generalizations of rates discussed previously[2] and have the forms

$$R_{12}^{(r)}(\Delta)=\frac{\Omega_r^2}{4}\frac{\Gamma_{12}^{od}}{\Delta^2+(\Gamma_{12}^{od}/2)^2}, \tag{2a}$$

$$R_{2c}^{(p)}=\sigma^{(p)}I^p. \tag{2b}$$

Here $\sigma^{(p)}$ is the p-photon off-resonance absorption cross section,[4] Ω_r is the r-photon Rabi frequency[5] for the 1-2 transition (Ω_r^2 is proportional to I^r), Δ is the effective detuning of r laser photons from the 1-2 transition frequency [$\Delta=\omega_{21}+sI-r\omega$, where sI is the linear (in intensity) Stark shift],[5] and Γ_{12}^{od} is the effective off-diagonal relaxation rate of the 1-2 transition. Then the $(r+p)$-photon ETL rate is given by

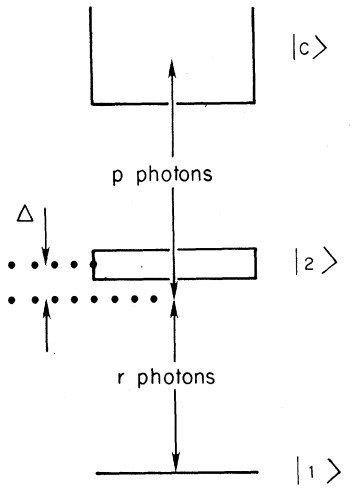

FIG. 1. Extended two-level (ETL) model atom. The first r photons are off resonance from the 1-2 transition by the small detuning Δ.

$$R_{\mathrm{ETL}}^{(r+p)}(\Delta)=\frac{\Omega_r^2}{4}\sigma^{(p)}I^p\frac{\Gamma_{12}^{od}/\Gamma_{12}^{d}}{(\omega_{21}+sI-r\omega)^2+(\Gamma_{12}^{od}/2)^2+\Omega_r^2\Gamma_{12}^{od}/2\Gamma_{12}^{d}}, \tag{3}$$

where Γ_{12}^{d} is the effective diagonal relaxation rate of the 1-2 transition. In a very general case one would have something like $\Gamma_{12}^{od}=A+\Gamma_{\mathrm{laser}}+R_{2c}^{(p)}+\Gamma_{\mathrm{coll}}^{od}$. That is, there would be contributions to Γ_{12}^{od} from the Einstein A coefficient for radiative decay from level $|2\rangle$, the laser bandwidth Γ_{laser}, the p-photon transition rate from $|2\rangle$ to $|c\rangle$, and the off-diagonal collision rate. Similarly one would have $\Gamma_{12}^{d}=A+R_{2c}^{(p)}+\Gamma_{\mathrm{coll}}$. Note that all of the parameters in $R_{\mathrm{ETL}}^{(r+p)}$ meet the simplicity requirement. They are all low-power quantities, not sensitive to resonance, and calculable via ordinary perturbation theory. Also, the appearance of s and $R_{2c}^{(p)}$ signal the implicit inclusion of more atomic levels than the two that are resonant; thus the term "extended" two-level model.

From expression (3) it is a simple matter to derive the nonlinear index K. I state explicitly here only the simplest case, appropriate to atomic beam experiments with narrow-band lasers ($\Gamma_{\mathrm{coll}}\cong\Gamma_{\mathrm{laser}}\cong 0$, $\Gamma_{12}^{od}=\Gamma_{12}^{d}$):

$$K_{\mathrm{ETL}}^{(r+p)}=r+p-\frac{2\Delta sI+\frac{1}{2}p(A+R_{2c}^{(p)})R_{2c}^{(p)}+\frac{1}{2}r\Omega_r^2}{\Delta^2+\frac{1}{4}(A+R_{2c}^{(p)})^2+\frac{1}{2}\Omega_r^2}. \tag{4}$$

K_{ETL} satisfies several analytic limiting conditions: (1) Far off resonance, $K\to r+p$; (2) far above saturation, when $\Omega_r\gg R_{2c}^{(p)}, A, \Delta$, then $K\to p$; (3) exactly on resonance with $R_{2c}^{(p)}\gg\Omega, A$, then $K\to r-p$. Both (1) and (2) are necessary features of any correct theory, and (3) has apparently been observed.[6] The resonance shape of K_{ETL} has a symmetric Lorentzian part and an antisymmetric dispersionlike part. It is the size of the transition's Stark-shift parameter s, relative to the width parameters A, $R_{2c}^{(p)}$, and Ω_r, that determines the shape of the sum of the parts. The maximum of K_{ETL} is not located either at the unperturbed resonance position $\Delta=sI$ or at the shifted resonance position $\Delta=0$. However, as

Mostowski has pointed out, the Lorentzian part of K_{ETL} must be negative, and so the center of gravity of $K_{ETL}(\Delta)$ must lie *below* the asymptotic value $r+p$, strongly at variance with published data.[6]

Of course it must be emphasized that the expressions for R and K given in (3) and (4) are model dependent. However, the model is analytically flexible, compatible with known physics in asymptotic limits, and conceptually quite natural. It is apparently the only model that is simple enough to yield explicit formulas for R and K, but complex enough to satisfy all five criteria (a)–(e) above. I believe it can provide specific and easily interpreted guidelines for future multiphoton work.

For comparison I show in Fig. 2 the degree of agreement that presently exists between the ETL model and multiphoton ionization experiments.[7] The best experimental data come from the work at Saclay on four-photon ionization of cesium.[6] I have superposed on the data points a curve showing K_{ETL} versus Δ. The curve was generated using (4) and the values of the required parameters given by Chang and Stehle,[8] except in the case of the Stark shift s, for which a measured value[6] was taken. The curve is plotted for an intensity of 4 GW/cm^2 and a Stark shift value of 4.3 cm^{-1}/GW cm^{-2}. The nominal intensity value in the experiments was approximately 1 GW/cm^2. The experimental points in the region where $K < 0$ are relatively unreliable.[9]

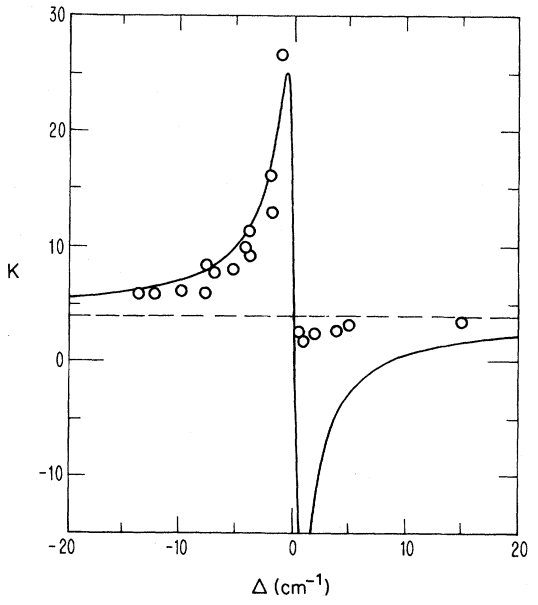

FIG. 2. The exponential index K for three-photon-resonant four-photon ionization of cesium. The circles represent the experimental data of Morellec, Normand, and Petite (Ref. 6) taken from Fontier and Trahin (Ref. 8). The solid lines are the ETL predictions for K for values of I and s (given in the text) in the neighborhood of the experimental values.

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Observation of Bound-Free-Bound Triplet Absorption Bands in Li_2

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Multiphoton ionization spectroscopy has yielded the first evidence for triplet absorption bands in Li_2 in the spectral region from 6215 to 5875 Å. This spectrum is the first of a new class of spectra resulting from a two-photon transition from a bound ($^3\Sigma_u^+$) van der Waals state to a previously unknown bound $^3\Lambda_u$ state (where $\Lambda = 0$, 1, or 2) through an intermediate free state ($^3\Pi_g$). The final state either autoionizes or collisionally forms the $^2\Sigma_g^+$ ground state of Li^{2+}.

The alkali-metal dimer triplet systems are of interest as potential excimer laser transitions since the lowest-lying triplet state, $^3\Sigma_u^+$, is very weakly bound.[1] Another point of possible practical as well as theoretical interest is that the lowest triplet state represents the interaction of spin-aligned lithium atoms, analagous to spin-aligned hydrogen atoms, a potential high-energy-density energy-storage medium.[2] Despite this interest, transitions between triplet states have either not been observed or not been analyzed because of experimental difficulties.[3] Lithium represents a special case since accurate *ab initio* calculations of its lowest-lying potential curves are available to aid in the analysis of new spectral features.[4-7] Correspondingly, these new calculations can be tested with new experimental results.

The techniques of absorption and fluorescence spectroscopy have yielded much information about the ground state, $^1\Sigma_g^+$, of the alkali metal dimers. However, the $^3\Sigma_u^+$ state has for the most part been inaccessible for spectroscopic investigation since the populations of the levels in this state are much smaller than those in the lower levels of the ground state. Moreover, even these lower levels, in transitions to higher electronic states, yield a confusion of overlapping bands.[8]

Two-photon spectra in alkali atoms and molecules have been detected via the method of ionization spectroscopy.[9,10] Since these spectra are detected only if excited-state product species are ionized, this method discriminates against the detection of absorptions to lower-lying energy levels. This detection method in its most sensitive variation, the space-charge ion detector, has been recently used in the investigation of photoionization of lithium vapor.[11] In a previous paper two of the present authors described both the method, "heat-pipe diode," and some preliminary results which established this new method's utility for the measurement of multiphoton ionization spectra.[12] Spectra from the red region, which will be discussed below, were not included in the earlier report.

An ionization spectrum from 5700 to 6300 Å is shown in Fig. 1(c). This was obtained with a heat-pipe diode at a lithium vapor pressure of 0.5 Torr with excitation by a chopped tunable dye laser (CR-599 operated broad band with <0.5 Å linewidth). The unsaturated signal of the molecular features depended upon the square of the light intensity indicating that two photons were absorbed to produce each ion. The atomic $2p$-to-$3d$ signal was more nearly a cubic than quadratic function of light intensity and was most probably due to the same ionization mechanism as the $2s$-to-$3d$ two-photon signal discussed previously.[11,12] For reference the neon optogalvanic