1 transition and \( \omega_1 + \omega_2 \) is slightly off resonance to the 0 \( \rightarrow \) 2 two-photon transition.\(^{1,2}\) the refractive index of the medium at \( \omega_2 \) will depend on the intensity of the field at \( \omega_1 \) and vice versa. Thus,

\[
\Delta n_{\text{NL}}(\omega_2) = \beta |E|^2
\]

(1)

and

\[
\Delta n_{\text{NL}}(\omega_1) = \beta_2 |E|^2.
\]

(2)

Under collision-free conditions \( \beta_1 = \beta_2 \), while under collision-dominated conditions \( |\beta_1| \gg |\beta_2| \). It can be shown that under collision-dominated conditions one can envisage practical applications of this effect in controlling the phase front of a laser beam at frequency \( \omega_2 \) by another laser at frequency \( \omega_1 \).

Refractive laser-controlled optical elements such as lenses and prisms can be made by appropriate tailoring of the spatial intensity distribution of the controlling laser. Detailed analysis shows that the power requirement of the controlling laser in this type of application is strongly dependent on the dimensions of the controlled beam. This is because in an ordinary refractive-lens application the phase lag in the center of the beam compared with that at the beam boundary is a quadratic function of the beam diameter. The power requirement is drastically reduced if, rather than an ordinary refractive lens or prism, one uses a Fresnel lens or a Fresnel prism.

Refractive laser-controlled optical elements for the laser beam at frequency \( \omega_2 \) can be made if the holographic image of the desired optical element is produced in the gas medium, by interfering two coherent laser beams at frequency \( \omega_1 \), with appropriate phase fronts. The spatial intensity modulation of the field at frequency \( \omega_1 \) will cause an identical spatial modulation of the refractive index of the gaseous medium at frequency \( \omega_2 \). For example, a volumetric planar grating for light at frequency \( \omega_2 \) can be generated by interfering two noncollinear plane waves at frequency \( \omega_1 \) in the medium.

Under collision-free conditions the real-time grating interaction is identical to the well-known phase-matched four-wave-mixing process. Therefore, under these conditions, the power requirement of the controlling laser is determined by the Manley–Rowe relations. On the other hand, in the collision-dominated regime the process deviates sharply from the four-wave mixing process, and the power requirement for the controlling laser becomes practically independent of the power of the controlled beam at \( \omega_2 \).

In the case of a laser-controlled holographic lens or mirror, the focusing efficiency depends on the extent to which the controlled beam at \( \lambda_2 \) satisfies the Bragg condition for the curved grating. When \( \lambda_2 \neq \lambda_1 \), the Bragg condition may still be well satisfied if inert gas is added so that

\[
\lambda_2 = \frac{n_0(\lambda_1)}{n_0(\lambda_2)} \lambda_1,
\]

(3)

where \( n_0(\lambda_1) \) is the linear refractive index of the medium at \( \lambda_1 \).

The response time of the real-time holographic optic described above is determined by the relevant lifetimes of the energy levels involved in the process and can be as short as 10 nsec. Diffusion in the gaseous medium causes a faster dissipation rate of the real-time grating and thereby increases the power required at \( \omega_1 \). On the other hand, diffusion also results in a decrease in the response time of the optical element. (12 min.)

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**WBB4. Polarization Renormalization due to Nonlinear Optical Generation, James J. Wynne, IBM T. J. Watson Research Center, P.O. Box 218, Yorktown Heights, New York 10598.**

Recent research has dealt with phenomena where coherent generation of electromagnetic (em) waves is intimately involved with multiphoton excitation, leading to results that cannot be explained by considering coherent generation and multiphoton excitation as separate phenomena. In particular, the disappearance with increasing vapor pressure\(^{1} \) and the reappearance with retroreflection\(^{2} \) of a multiphoton ionization signal, as well as the disappearance with increasing pressure of fluorescence from a multiphoton excited state,\(^{3} \) have been explained by considering the effect of coherent third-harmonic generation on the multiphoton excitation.\(^{4,5} \) These explanations present specific calculations showing that the total excitation is reduced by the harmonic-generation process, but they fail to address the underlying general principle that leads to such a reduction.

I show that this reduction is dictated by the laws of em-wave propagation, namely, the em-wave equation that is derived from Maxwell's equations. I treat the problem entirely classically, making no recourse to a quantum-mechanical description of the linear or nonlinear system response. The key idea is to consider the total electric polarization. This is the sum of the nonlinear source polarization (the driving force of generation) and the linear polarization, i.e., the linear response of the polarizable medium to the generated electric field. Then, when the coherently generated em wave experiences linear absorption, leading to spatial decay of free waves at the frequency of the generated light so that the spatially persisting wave is described solely by a driven wave, careful consideration of the relationship between the nonlinear source polarization and the linear polarization is sufficient to explain the anomalous experimental results.\(^{1-3} \) In effect, the nonlinear source polarization is renormalized to a new value by the nonlinear optical generation of an electric field and its accompanying linear polarization. (12 min.)


**WBB5. Amplitude- and Frequency-Modulation Raman Heterodyne Detection of Optical Double Resonance, J. Miynek, Chr. Tamm, E. Buhr, and N. C. Wongs.**

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Recently a simple transmission method for phase-sensitive optical heterodyne detection of \( \tau_1 \)-induced sublevel coherence was successfully applied in dilute solids at low temperature.\(^{1,2} \) The technique extends previous methods in optical-pumping double resonance\(^{3} \) and relies on a coherent Raman process being stimulated by a resonant pulsed or cw radio-frequency field and an optical field. It has been predicted\(^{4} \) that the corresponding Raman heterodyne signal generally contains amplitude-modulation (AM) and frequency-modulation (FM) sideband contributions, where the AM is supposed to dominate in the optically resonant case and the FM in the nonresonant case. Here we demonstrate that in an atomic vapor, a simple

![Fig. WBB5-1](image-url)
MAGNETIC FIELD (mT)

Fig. WBB5-2. Raman heterodyne signal of Zeeman coherences for the $\lambda = 570.6 \text{ nm}$ $^2F_1 \rightarrow ^2F_0$ transition showing the in-phase and quadrature output of the lock-in amplifier. The rf frequency is kept fixed ($\omega = 2\pi \times 8.7 \text{ MHz}$) and the $B_0$ field is scanned (Larmor frequency: $2\pi \times B_0 \times 21 \text{ MHz/mT}$). a) RF amplitude $B_1 = 1.6 \mu\text{T}$; b) $B_1 = 50 \mu\text{T}$.

optical-phase-shift technique allows the separate study and comparison of both coherent Raman signals.

Experiments were performed on Zeeman sublevels in atomic samarium vapor using simple $J = 1 \rightarrow J' = 0$ [see Fig. WBB5-1(b)] or $J = 0 \rightarrow J' = 1$ transitions; in this summary we only discuss the Sm $570.6\text{ nm}^2F_1 \rightarrow ^2F_0$ transition. A schematic diagram of the apparatus is shown in Fig. WBB5-1(a). The laser field $E_L$ of frequency $\Omega$ is polarized parallel to the transverse static $B_0$ field ($P\parallel B_0$); it only drives the $x$ transition and thus produces an alignment in the ground-state $m$ sublevels. A longitudinal rf field of frequency $\omega$ resonantly excites $|\Delta m| = 1$ coherences and the simultaneous presence of the light field $E_L$ gives rise to coherent Raman sidebands $E_s$ with frequencies $\Omega \pm \omega$ and polarization states $\sigma_a$ [Fig. WBB5-1(c)]. A polarization analyzer $A$ is used to project the carrier ($\Omega$) and the sidebands along a common direction [Fig. WBB5-1(d)], and a photodiode detects the corresponding optical beat signal at frequency $\omega = |\Omega - (\Omega \pm \omega)|$. In this configuration, only the AM is detected and the FM cancels out. A high-frequency lock-in amplifier is used for narrow-bandwidth detection of this heterodyne signal; its output yields absorption- or dispersion-type line shapes as shown in Fig. WBB5-2. The splitting of the Raman signal at high rf amplitude stems from two-photon resonant rf transitions between equally spaced $m$ sublevels; it is in good agreement with a four-level calculation for the coherent Raman process.

For our special geometry, the FM instead of the AM signal contributions [see Fig. WBB5-1(d)] can easily be detected by using an optimal $\lambda/4$ plate in front of the analyzer to shift the phase of the carrier $E_L$ by $90^\circ$. In Fig. WBB5-3 typical results are shown for the AM- and FM-detection modes under otherwise unchanged experimental conditions. Our measurements clearly demonstrate that the amplitude and shape of the AM and FM signals can have a significantly different dependence on, e.g., buffer-gas pressure, laser detuning, and optical power. At present, the development of a modified theory for the optical interaction, including optical hole-burning effects and a detailed treatment of relaxation processes, is in progress to understand the various features and differences in AM- and FM-Raman heterodyne signals.

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WCCI. Spectroscopy of the Rydberg Molecules $^2H_2$, $^2D_2$, $^2D_3$, and $^2D_4$, H. Figger, Y. Fukuda, W. Ketterle, and W. Schreppe, Max-Planck-Institut für Quantenoptik, D-8046 Garching, Federal Republic of Germany, J. K. G. Watson, Herzbberg Institute of Astrophysics, National Research Council of Canada, Ottawa, Canada K1A0R6, and H. Walther, Max-Planck-Institut für Quantenoptik and Sektion Physik, Universität München, D-8046 Garching, Federal Republic of Germany.

In 1979 the optical emission spectra of $^2H_2$ and $^2D_2$ were discovered in hollow-cathode discharges by G. Herzberg and co-workers. According to this work, $^2H_2$ and $^2D_2$ have to be considered as Rydberg molecules with one electron orbiting around a $K^+$ or $K^+$ core and with a dissociating ground state. We generated molecular beams of $^2H_2$, $^2D_2$, and $^2D_4$ in Rydberg states by neutralizing the corresponding mass-selected positive-ion beams in an alkali-vapor cell. Behind the cell the characteristic bands around 5600, 5800, 6055, and 7100 Å were observed. Examples are given for $^2D_3$ in Fig. WCCI-1 and for $^2D_4$ in Fig. WCCI-2. So far, only this beam setup allowed us to identify spectra of the mixed isotopes. In contrast to the spectra emitted by a hollow-cathode discharge in hydrogen they are free from lines of diatomic or the tristatic hydrogen molecules.

All bands observed correspond to transitions between electronic states with main quantum numbers $n = 3$ and $n = 2$, the latter one is predissociating and therefore responsible for the linewidths of a few angstroms. Transitions from Rydberg states with $n > 3$ to $n = 2$ states were not yet observed.

![Fig. WCCI-1. Spectrum around 5600 Å emitted by $^2D_3$ in a molecular beam.](image_url)