

## Linear and Nonlinear Optical Measurements of the Lorentz Local Field

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We have studied the resonant optical response of a dense atomic potassium vapor under conditions such that the response is strongly influenced by local-field effects. Our experimental results are in good agreement with the predictions of theory. We have also discovered a collision-induced shift of the potassium resonance lines given by  $\Delta\omega_{\text{col}} = \beta N$ , where  $\beta = -5.0 \times 10^{-8} \text{ sec}^{-1} \text{ cm}^3$  for the  $4^2S_{1/2} \leftrightarrow 4^2P_{1/2}$  transition and where  $\beta = -3.0 \times 10^{-8} \text{ sec}^{-1} \text{ cm}^3$  for the  $4^2S_{1/2} \leftrightarrow 4^2P_{3/2}$  transition.

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One of the fundamental problems of optical physics concerns the proper procedure for relating macroscopic properties of material systems, such as susceptibilities and refractive indices, to microscopic properties, such as atomic and molecular polarizabilities [1-3]. The proper treatment of this problem is extremely difficult, because in principle each atom of the sample interacts with every other atom, and all of these interactions must be accounted for in describing the macroscopic properties of the material. One of the standard but approximate procedures for accounting for these interactions is to introduce the concept of the local field  $\mathbf{E}_{\text{loc}}$ . The local field is taken to be the effective field, due both to external sources and to all other atoms of the material sample, that acts on a typical atom of the material. The local field is to be distinguished both from the microscopic electric field and from the macroscopic field  $\mathbf{E}$ , which is the averaged microscopic field and which is the field that appears in the macroscopic Maxwell equations. According to a prescription described by Lorentz [4], the local and macroscopic fields within a isotropic environment are related (in Gaussian units) by

$$\mathbf{E}_{\text{loc}} = \mathbf{E} + (4\pi/3)\mathbf{P}, \quad (1)$$

where  $\mathbf{P}$  denotes the polarization of the material system. For the case of a material with a purely linear response, the polarization can be taken to have the form  $\mathbf{P} = N\alpha\mathbf{E}_{\text{loc}}$ , where  $N$  represents the number density of atoms and  $\alpha$  represents the atomic polarizability. If we define the dielectric constant in the conventional manner so that  $\epsilon = 1 + 4\pi\chi$ , where the linear susceptibility is defined by the relation  $\mathbf{P} = \chi\mathbf{E}$ , we find from Eq. (1) that  $\epsilon$  satisfies the well-known Lorentz-Lorenz law [4]

$$\frac{\epsilon - 1}{\epsilon + 2} = \frac{4\pi}{3} N\alpha. \quad (2)$$

The Lorentz-Lorenz law can be written alternatively as  $\epsilon = 1 + 4\pi L N\alpha$ , where the local-field enhancement factor  $L = (\epsilon + 2)/3$  can be interpreted as the factor by which the material response  $\epsilon - 1$  exceeds the predicted value when calculated in the absence of local-field effects.

Local-field effects also play an important role in determining the magnitude of the nonlinear response of optical materials [5]. For example, within the above framework, Bloembergen [6] has shown that the third-order susceptibility  $\chi^{(3)}(\omega_4; \omega_3, \omega_2, \omega_1)$  of a collection of polarizable molecules is given by

$$\chi^{(3)} = L(\omega_4)L(\omega_3)L(\omega_2)L(\omega_1)N\gamma^{(3)}, \quad (3)$$

where  $\gamma^{(3)}(\omega_4; \omega_3, \omega_2, \omega_1)$  is the molecular hyperpolarizability. Note that  $L$  appears to fourth order in the expression; local-field effects play an even more important role in determining the susceptibilities of higher-order nonlinear optical interactions. Moreover, local-field effects play a crucial role in determining the nonlinear response of composite optical materials [7]. For example, large enhancements in the nonlinear optical response of semiconductor-doped glasses [8] and of metal colloids [9] have been reported. Because of the importance of local-field effects in determining the magnitude of the linear and nonlinear optical response, and due to the approximate character of the prescriptions given by Eqs. (1)-(3) for treating local-field effects, we have undertaken an experimental investigation of local-field effects in linear and nonlinear optics.

In this Letter, we report the results of our experimental study of local-field effects in determining the optical response of a dense atomic vapor. We have chosen an atomic vapor for use in this study because explicit expressions for the linear polarizability  $\alpha$  and hyperpolarizability  $\gamma^{(3)}$  are available and because the validity of Eqs. (2) and (3) has not previously been examined for resonant excitation of an atomic system. In fact, it is precisely under conditions of resonant excitation where the interaction of neighboring atoms becomes strongest and where the validity of the prescriptions given by Eqs. (2) and (3) for treating local-field effects becomes most questionable. When the standard expression

$$\alpha = \frac{f r_e \lambda_0 c / 4\pi}{\omega_0 + \Delta\omega_{\text{col}} - \omega - i\gamma_2} \quad (4)$$

for the linear polarizability of an isolated optical transi-

tion of oscillator strength  $f$ , transition frequency  $\omega_0$ , and vacuum transition wavelength  $\lambda_0$  is introduced into the Lorentz-Lorenz relation (2), we find that the dielectric constant is given by the expression [4]

$$\epsilon = 1 + \frac{fr_e\lambda_0cN}{\omega_0 + \Delta\omega_{\text{col}} + \Delta\omega_L - \omega - i\gamma_2}, \quad (5)$$

where the quantity  $\Delta\omega_L = -\frac{1}{3}fr_e\lambda_0cN$  is the Lorentz redshift and  $r_e = e^2/mc^2$  is the classical electron radius. Comparison of Eqs. (4) and (5) shows that the influence of local-field effects is to shift the resonance of the dielectric constant from that of the atomic polarizability by the amount  $\Delta\omega_L$ . Friedberg, Hartmann, and Manassah [10] have shown that under conditions of nonlinear response the value of the Lorentz shift depends upon the degree of population inversion between the two interacting levels. The implications of this inversion-dependent frequency shift on intrinsic optical bistability [11] and on the propagation of optical solitons [12] have been examined.

Our first experiment involves measurement of the resonant structure of  $\epsilon(\omega)$  for various values of the number density  $N$  of an atomic potassium vapor. The position and shape of this resonance also depend on any possible shift  $\Delta\omega_{\text{col}}$  in the atomic resonance frequency due to collisional effects and on any collisional contribution to the atomic linewidth  $\gamma_2$ . According to standard line-broadening theory [13], the atomic linewidth can be represented as  $\gamma_2 = \frac{1}{2}\gamma_{\text{nat}} + \gamma_{\text{self}}$ , where  $\gamma_{\text{nat}}$  represents the natural (i.e., radiative) linewidth and where the contribution to the atomic linewidth due to collisions between potassium atoms is given by

$$\gamma_{\text{self}} = fr_e\lambda_0cN[(2J_g + 1)/(2J_e + 1)]^{1/2},$$

where  $J_g$  and  $J_e$  are the angular momentum quantum numbers of the ground and excited states, respectively. According to Eq. (5), experiments performed in the linear regime cannot distinguish between the Lorentz shift  $\Delta\omega_L$  and the collisional shift  $\Delta\omega_{\text{col}}$ . However, as we show below, these two contributions are not simply additive quantities in the expression for the nonlinear response, and consequently through comparison of our linear and nonlinear measurements we are able to determine these contributions separately.

For an atomic vapor with a number density sufficiently large that local-field effects are important, the penetration depth of resonant light into the vapor is of the order of  $\lambda_0/2\pi$ , and consequently the vapor is essentially opaque for the transmission of resonant radiation. For this reason, we have studied the resonant structure of the dielectric constant by measuring the spectrum of the reflectivity of the interface between a dense potassium vapor and a sapphire plate that forms the window of the atomic vapor cell [14,15]. Under our experimental conditions, the mean interatomic separation was much smaller than the penetration depth of light into the medium, and, for this reason, our measured reflectivity signal was not

significantly affected by atoms whose properties were strongly modified by the surface of the sapphire window. The highly wavelength-dependent reflectivity that occurs near an atomic resonance is known as selective reflection and has been studied by many workers [16-18]. However, these earlier studies of selective reflection were performed either using number densities sufficiently small that local-field effects were unimportant or using nonlaser sources with insufficient spectral resolution to be sensitive to local-field effects.

Some of our experimental results for the  $4^2S_{1/2} \leftrightarrow 4^2P_{1/2}$  transition are shown in Fig. 1(a). Here the reflectivity of the interface is plotted as a function of laser frequency for several different values of the atomic number density. Note that the spectral variation in reflectivity becomes more pronounced and that the reflectivity curves shift to lower frequency as the number density is increased. The observed shift is direct evidence of the Lorentz redshift, which is a manifestation of local-field effects. Figure 1(b) shows the theoretically predicted values of the reflectivity, which exhibit the same behavior as the experimental curves. The theoretical curves were calculated using the conventional expression for the Fresnel reflection coefficient and the tabulated values of the refractive indices of sapphire, and by performing a Doppler average of the dielectric constant of potassium as predicted by Eq. (5) with  $\Delta\omega_{\text{col}}$  given by a formula described below.

The results of Fig. 1 are analyzed more quantitatively in Fig. 2. Here part (a) gives the density dependence of the width  $\gamma_{\text{SR}}$  of the selective reflection feature, which is taken to be the difference between the frequency of highest reflectivity and that of lowest reflectivity; and part (b) gives the density dependence of the shift  $\Delta\omega_{\text{SR}}$  of the selective reflection feature, which is taken to be the difference between the average of the frequencies of maximum and minimum reflectivities and the resonance frequency of an isolated atom. This shift is of the order of the shift in  $\epsilon$  (that is,  $\Delta\omega_{\text{col}} + \Delta\omega_L$ ), but is not exactly equal to the shift in  $\epsilon$  because the surface reflectivity depends in a nonlinear manner on  $\epsilon$ . The results for the width [part (a)] are in excellent agreement with the pre-

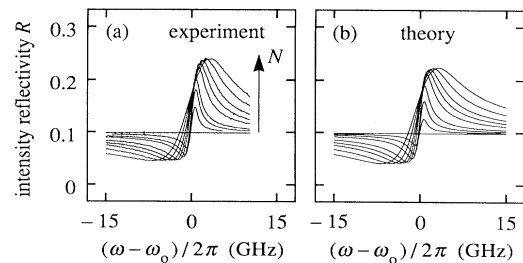


FIG. 1. Frequency dependence of the reflectivity of the interface between a sapphire window and a potassium vapor for various values of the atomic number density, for frequencies near the  $4^2S_{1/2} \leftrightarrow 4^2P_{1/2}$  transition.

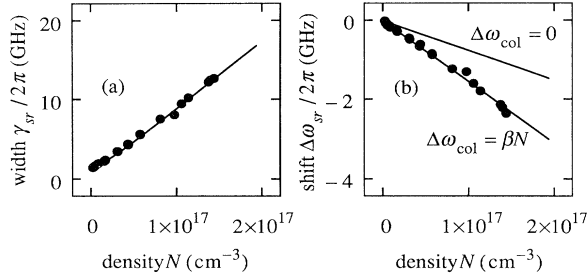


FIG. 2. (a) Width  $\gamma_{SR}$  and (b) spectral shift  $\Delta\omega_{SR}$  of the selective reflection features of Fig. 1 plotted as functions of the atomic number density. The lines give the predictions of theory. For (b), good agreement is obtained under the assumption that  $\Delta\omega_{col} = \beta N$ , where  $\beta = -5.0 \times 10^{-8} \text{ sec}^{-1} \text{ cm}^3$ .

dictions based on standard line-broadening theory, as outlined above. However, we see from part (b) of the figure that the shift is somewhat greater than that predicted solely on the basis of the Lorentz shift and that agreement between theory and experiment is obtained if we assume that there is a collisional contribution to the frequency shift given by  $\Delta\omega_{col} = \beta N$  with  $\beta = -5.0 \times 10^{-8} \text{ sec}^{-1} \text{ cm}^3$ .

Our second experiment measures the nonlinear optical response by surface phase conjugation [19], which involves the interaction of a weak signal wave with a pump wave that is normally incident on the interface between the sapphire window and the potassium vapor. The interference of these waves produces a spatially varying intensity pattern on the surface which leads to a spatial variation in the Fresnel reflection coefficient; the reflection of the pump wave from this reflectivity distribution then creates the phase conjugate of the signal wave. Figure 3 shows the intensity of the phase-conjugate signal as a function of the laser frequency for several different values of the atomic number density. The experimental results shown in part (a) of the figure are in excellent agreement with the theoretical results shown in part (b). The theoretical predictions are obtained from a generalization of the established theory of surface phase conjugation, which yields the prediction that the phase-conjugate intensity reflectivity for an *s*-polarized probe wave incident at an angle of  $\theta_s$  is given by [20]

$$R_{PC} = \frac{36\pi^4}{c^2 \epsilon_s} \frac{|t_{sr}|^2 |t_{vs}|^2 |t_{sr}^\perp|^4}{|\zeta|^2 |\sqrt{\epsilon} + i \text{Im}(\zeta)|^2} I_{\text{pump}} |\chi^{(3)}|^2, \quad (6)$$

where  $I_{\text{pump}}$  is the intensity of the pump wave. The quantities  $t_{sr}$ ,  $t_{vs}$ , and  $t_{sr}^\perp$  are Fresnel amplitude transmission coefficients and  $\zeta = [\epsilon - \epsilon_s \sin^2(\theta_s)]^{1/2}$ , where  $\epsilon_s$  is the dielectric constant of sapphire. The third-order susceptibility that appears in expression (6) is given, including local-field corrections according to the prescription of Eq. (3), as

$$\chi^{(3)} = L^2 |L|^2 \frac{f r_e \lambda_0 c N}{12 \pi \gamma_2 |E_s|^2} \frac{\delta - i}{(1 + \delta^2)^2}, \quad (7)$$

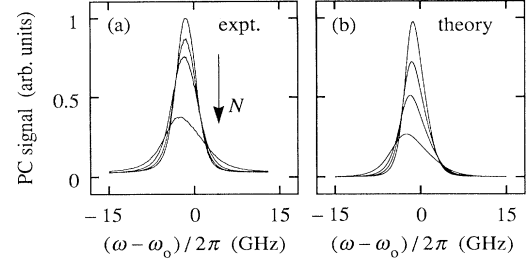


FIG. 3. Intensity of the phase-conjugate signal plotted as a function of detuning from the  $4^2S_{1/2} \leftrightarrow 4^2P_{1/2}$  atomic resonance frequency for various values of the atomic number density.

where  $\delta = (\omega - \omega_0 - \Delta\omega_{col})/\gamma_2$  and  $L = [\epsilon(\omega) + 2]/3$ . We see from Fig. 3 that the maximum phase-conjugate reflectivity decreases as the number density is increased, owing to an increase in the saturation parameter  $|E_s|^2 = \gamma_{\text{nat}} \gamma_2 h / 2 f r_e \lambda_0 c$  with increasing values of  $\gamma_2$ , and that the curves shift to lower frequency. The magnitude of this shift  $\Delta\omega_{PC}$  is shown as a function of atomic number density in Fig. 4, and is seen to be in excellent agreement with the predictions of our theoretical model, in which we set  $\Delta\omega_{col} = \beta N$  with  $\beta$  equal to the same value ( $-5.0 \times 10^{-8} \text{ sec}^{-1} \text{ cm}^3$ ) obtained from the linear response measurements shown in Fig. 1. We also show as dashed lines in Fig. 4 the prediction of Eq. (6) under the artificial assumptions that (i) all of the shift observed in the linear response experiment was due to local-field effects (i.e., that  $\beta$  vanishes but that  $\Delta\omega_L = -0.56 f r_e \lambda_0 c N$  rather than the Lorentz prediction of  $-0.33 f r_e \lambda_0 c N$ ) and (ii) that all of the shift was due to a collisional shift (i.e., that  $\Delta\omega_L = 0$  but that  $\beta = -1.2 \times 10^{-7} \text{ sec}^{-1} \text{ cm}^3$ ). Under any of these sets of assumptions, the total predicted shift in the linear response is equal to that observed in the linear response experiment, but we see that only the Lorentz prediction along with a collision shift with

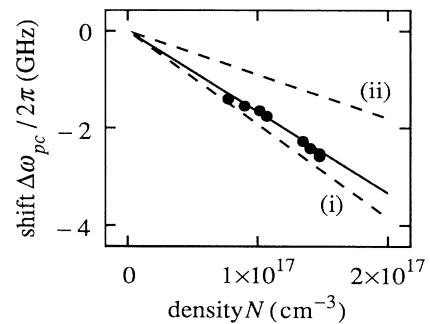


FIG. 4. Spectral shift  $\Delta\omega_{PC}$  of the maximum of the tuning curve for surface phase conjugation from Fig. 3 plotted as a function of the atomic number density. The solid line gives the prediction of theory based on the Lorentz local field and a collision shift given by  $\beta = -5.0 \times 10^{-8} \text{ sec}^{-1} \text{ cm}^3$ . The disagreement with the dashed curves rules out other possible models that are described in the text.

TABLE I. The measured value of  $\beta$  and the value of  $f$  and  $\lambda_0$  from Ref. [22] for the doublet lines of potassium.

Atomic transition	$\beta$ (sec <sup>-1</sup> cm <sup>3</sup> )	$f$	$\lambda_0$ (nm)
$4^2S_{1/2} \leftrightarrow 4^2P_{1/2}$	$-5.0 \times 10^{-8}$	0.339	770.1
$4^2S_{1/2} \leftrightarrow 4^2P_{3/2}$	$-3.0 \times 10^{-8}$	0.682	766.7

$\beta = -5.0 \times 10^{-8}$  sec<sup>-1</sup> cm<sup>3</sup> is in agreement with both the linear and the nonlinear optical data.

Collisional shifts of the atomic resonance frequency have not previously been reported for the resonance lines of alkali vapors and have been speculated to vanish [13]. However, the magnitude of the shift that we have measured is probably too small to have been seen in previous experiments. We believe that this shift can be explained at least in part in terms of the van der Waals interaction of two separated potassium atoms [21]. A simple, order-of-magnitude calculation shows that the transition frequency will be shifted to lower frequencies due to this effect by an amount of the order of  $4\pi^2 Ne^2 a_0^2 / 3h$ , where  $a_0$  is the Bohr radius, which corresponds to a shift of 1 GHz at a number density  $N$  of  $1 \times 10^{17}$  cm<sup>-3</sup>. The interaction energy due to the overlap of atomic wave functions during collisions with small impact parameter is also expected to make a contribution.

We have also studied the linear and nonlinear optical response associated with the  $4^2S_{1/2} \leftrightarrow 4^2P_{3/2}$  transition of potassium. Our results here are also in good agreement with the predictions of theory. The results for the two resonance lines are summarized in Table I.

In conclusion, we have measured the resonant linear and nonlinear optical responses of a dense atomic potassium vapor under conditions such that local-field effects played a large role in determining the magnitude of the response. Our results are in agreement with the predictions of the Lorentz-Lorenz law in the linear case and with Bloembergen's prediction of the local-field correction to the third-order susceptibility for the nonlinear case, once we include a collision-induced shift of the potassium resonance frequency.

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