Nonlinear Generation of 104.8-nm Radiation within an Absorption Window in Zinc

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Two autoionizing levels which are separated by a few decay widths may exhibit a sharp interference or window in their absorption profile and also make canceling contributions to the refractive index at the absorption minimum. A correct choice of intermediate mixing levels prevents a similar cancellation in the nonlinear susceptibility. Using UV lasers with energies of about mJ and pulse lengths of 5 ns, we generate 0.23 μJ per pulse at 104.8 nm.

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On occasion two atomic energy levels which lie above an ionization potential and decay by autoionization to the same continuum may exhibit a strong destructive interference in their absorption profile. In this Letter we show how, by the correct choice of intermediate mixing levels, one may use such a window to generate sum-frequency radiation.

Before proceeding we note that Armstrong and Wynne[1] have shown that the profiles for absorption and for the nonlinear susceptibility $\chi^{(3)}$ in the vicinity of a single autoionizing resonance need not be the same. By utilizing the interference of two lines, rather than that of a single line and a continuum, this work extends theirs in two ways: First, the interference of two lines results in a near zero in the contribution of the lines to the refractive index at the generated frequency, as well as to the absorption. This is not an accident and will occur whenever autoionization is determined by the same dipole matrix elements that determine the absorption. Second, because the interference between two lines is much steeper than that of a single line and a continuum, it occurs closer to both lines and results in a much greater increase in the nonlinearity length product.

The connection to the suggestion of Harris, Field, and Imamoğlu[2] and to the recent experiments of Hakuta, Marmet, and Stoicheff[3] is also to be noted: By applying an electromagnetic field to mix a (perfectly) metastable level with a lifetime-broadened level, one may create (in the absence of Doppler and collision broadening) the ideal situation of zero loss, zero contribution to the refractive index, and constructive interference in the nonlinearity. This work does not quite accomplish these goals: Here, the nonlinear susceptibility and the small but nonzero linear susceptibility are the result of the departure from L-S coupling.

Figure 1 shows a partial energy-level diagram of neutral Zn. The interfering levels $3d^94s^24p\,^1P_1^\circ$ and $3d^94s^24p\,^3D_1^\circ$ decay primarily by autoionization to a common $\mathrm{Zn}(3d^{10}4s\,^2S_{1/2})$ ion and a $^1P_1$ electron. Figure 2 shows the absorption cross section as a function of photon frequency, as obtained from the data of Marr and Austin.[4] The absorption window near 95 370 cm^{-1}

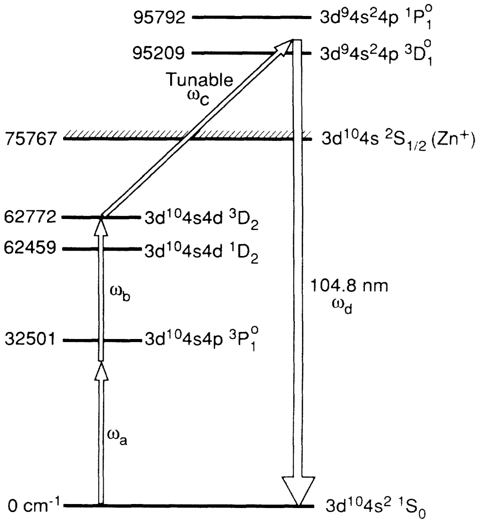

FIG. 1. Partial energy-level diagram of neutral zinc.

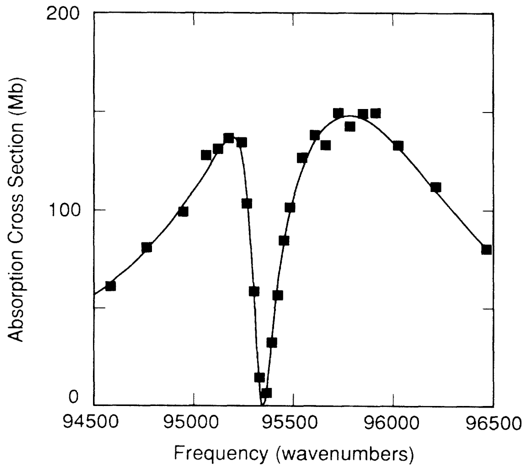

FIG. 2. Absorption cross section as a function of frequency. Data points are from Marr and Austin (Ref. 4). The solid curve is a fit using Eq. (6) from Harris (Ref. 5).

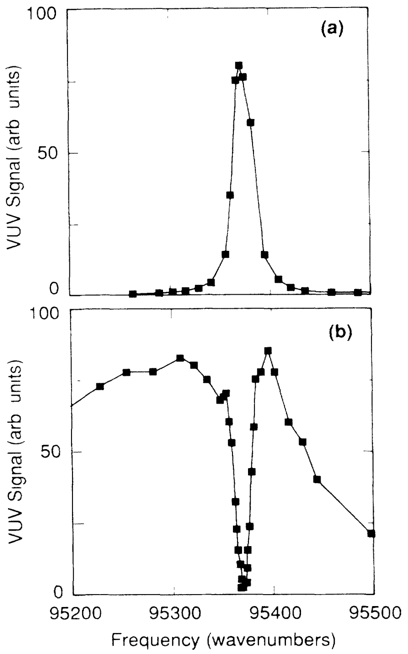

FIG. 3. Generated VUV signal as a function of ω_d with the two-photon-resonant intermediate level (a) $3d^{10}4s4d\,^3D_2$ and (b) $3d^{10}4s4d\,^1D_2$.

results primarily from the interference of two paths (to the end product of the autoionizing decay) which have equal magnitude and opposite sign. The interference is sufficiently strong so as to decrease the absorption, as compared to that at the peak of the resonance, by a factor of about 120.

Two experiments whose purpose was to determine the behavior of the nonlinearity length product in the vicinity of the absorption window were performed. In the first of these, a laser with a frequency $\omega_a = \omega_b$ was tuned to be two-photon resonant[6] with the triplet intermediary $3d^{10}4s4d\,^3D_2$ (Fig. 1). In the second, this laser was tuned to be two-photon resonant with the singlet intermediary $3d^{10}4s4d\,^1D_2$. In both experiments, the laser with frequency ω_c was tuned through the absorption window.

The lasers were focused into a heat-pipe cell with a 50-cm lens to a diameter of about 200 μm. To allow the Zn to recirculate on the wick of the heat pipe, Li was added in a molar ratio of Zn:Li of 1:3. The density of the zinc vapor was measured to be 5×10^{16} cm^{-3} over a 10-cm-long region. The beams at ω_a, ω_b, and ω_c were produced by frequency doubling the output of Nd-doped yttrium-aluminum-garnet-pumped dye lasers operated at a repetition rate of 10 Hz. The vacuum-ultraviolet (VUV) radiation near 104.8 nm was collected by a 0.2-m monochromator (Acton Research Corporation) and detected by a microchannel plate, and the detected signal was averaged with a boxcar integrator. Absolute energies in the VUV were determined by a calibrated vacuum photodiode placed after the monochromator and were corrected for the monochromator throughput.[7]

TABLE I. Matrix elements for calculation of linear and nonlinear susceptibilities.

	Matrix elements (a.u.)		
	$3d^{10}4s^2\,^1S_0$	$3d^{10}4s4d\,^1D_2$	$3d^{10}4s4d\,^3D_2$
$3d^{10}4s4p\,^3P_1^\circ$	−0.0271	−0.0188	−1.304
$3d^{10}4s4p\,^1P_1^\circ$	−2.45	−2.73	0.001 88
$3d^94s^24p\,^3D_1^\circ$	0.3753	−0.061	−0.0119
$3d^94s^24p\,^1P_1^\circ$	−0.8989	0.143	0.005 15

Figures 3(a) and 3(b) show the VUV signal as a function of the generated frequency, ω_d, using the triplet and singlet intermediaries, respectively. In the case of the triplet intermediary, a sharp peak is observed at the wavelength of the absorption window. For the singlet intermediary, a sharp dip is obtained at the same wavelength.

When using the triplet intermediary, with laser energies of 0.3 mJ at $\omega_a = \omega_b$ and ω_c, the generated 104.8-nm energy at the output of the zinc cell was 0.17 nJ per pulse. The conversion efficiency (generated VUV energy per total laser energy) was therefore 3×10^{-7}. To improve the conversion efficiency, another tunable laser was used to reduce the detuning of ω_a from the dominant first intermediate level $3d^{10}4s4p\,^3P_1^\circ$, and the energy of ω_c was raised to 2 mJ. In this case, the generated 104.8-nm energy was 0.23 μJ per pulse, implying a conversion efficiency of 9×10^{-5}. (Here, the conversion efficiency from the weakest beam ω_a to 104.8 nm was 1×10^{-3}.) The coherence length (calculated) was limited by the dispersion of the UV laser beams to 1.5 cm, and no attempt was made to correct this phase mismatch.

The above results may be understood by reference to the eigenfunction expansion of the autoionizing levels given below, to Table I which gives the matrix elements which are pertinent to this experiment, and to Table II which gives the partial and total decay rates of each of the autoionizing levels. These quantities are obtained from the atomic physics code RCN/RCG.[8]

A partial expansion of the autoionizing levels is

$$3d^94s^24p\,^1P_1 = 0.88\,^1P_1 + 0.41\,^3D_1 - 0.13\,^3P_1 + \cdots + ,$$
$$3d^94s^24p\,^3D_1 = -0.36\,^1P_1 + 0.89\,^3D_1 + 0.24\,^3P_1 + \cdots + . \tag{1}$$

TABLE II. Calculated autoionization rates.

	Autoionization rate (sec^{-1})	
Decay channel	$3d^94s^24p\,^3D_1$	$3d^94s^24p\,^1P_1$
$^3P_1$ continuum	1.50×10^{12}	$5.65\ \times10^{11}$
$^1P_1$ continuum	7.50×10^{13}	4.294×10^{14}
Total	7.65×10^{13}	4.300×10^{14}

Since the (pure L-S) $^3D_1$ component is of odd parity and even angular momentum, it may not decay by autoionization.[9] Therefore, to first order, both levels decay to the $^1P_1$ continuum. It is for this reason that the interference is so pronounced. The much slower decay of these levels to the $^3P_1$ continuum causes a (calculated) absorption minimum of about 1.3 Mb, which is too small to be noted in the Marr and Austin data of Fig. 2. If both the direct photoionization channel from ground and the decay to the $^3P_1$ continuum are neglected, then a perfect interference (absorption zero) occurs at the point between the two autoionizing levels where their virtual contributions have equal magnitude and opposite sign. If μ_{gi}, Γ_i, and $\Delta\omega_i$ denote the matrix element to the ground level, decay rate to the $^1P_1$ continuum, and frequency detuning from each of the (ith) autoionizing levels, then the zero occurs when

$$\Delta\omega_1\mu_{g2}\Gamma_2^{1/2}=\Delta\omega_2\mu_{g1}\Gamma_1^{1/2}\,. \tag{2}$$

With these same approximations, the contribution of the autoionizing levels to the refractive index will have a zero when

$$-4\Delta\omega_1\Delta\omega_2(\Delta\omega_2\mu_{g1}^2+\Delta\omega_1\mu_{g2}^2) =(\Gamma_1\Delta\omega_2+\Gamma_2\Delta\omega_1)(\Gamma_2^{1/2}\mu_{g1}+\Gamma_1^{1/2}\mu_{g2})^2\,. \tag{3}$$

Therefore, if the ratio of the matrix elements squared is equal to the ratio of the autoionizing rates, there will be a zero in the contribution to the refractive index at exactly the frequency of the absorption zero. From Tables I and II we find that $(\mu_{g2}^2/\mu_{g1}^2)/(\Gamma_2/\Gamma_1)=1.002$. (This occurs because a $4p$ electron makes a dipole transition to a $3d$ orbit and a $4s$ electron is ejected during autoionization.) Thus, to a good approximation, a zero in the contribution to the refractive index coincides with the absorption minimum. At the position of the absorption minimum and at the operating density, a more complete calculation gives a contribution of the two autoionizing lines to the refractive index of 5×10^{-9}.

We next consider the nonlinearity which governs the sum-frequency process. Figures 4(a) and 4(b) show the calculated nonlinear susceptibility when using the triplet $3d^{10}4s4d\,^3D_2$ and singlet $3d^{10}4s4d\,^1D_2$ intermediaries, respectively. If the two-photon perturbation path is through the triplet intermediary, then the frequency ω_c excites the $^3P_1$ and $^3D_1$ components of the interfering autoionizing levels. The excitation occurs primarily through small components of the $3d^{10}4snp$ and $3d^94s4p4d$ configurations, which are not shown in the expansion of Eq. (1). If the two-photon perturbation path is through the singlet intermediary $3d^{10}4s4d\,^1D_2$, then the excitation of the autoionizing levels occurs through the $^1P_1$ terms of these additional configurations. The amplitudes of these components have the same ratio as the primary $3d^94s^24p\,^1P_1$ components shown in Eq. (1). In this case $\chi^{(3)}$ has a zero at the same frequency as the real and imaginary parts of $\chi^{(1)}$.

In summary, we have shown an example of the use of an absorption window to substantially increase the nonlinearity length product of an otherwise opaque nonlinear media. This is done by exploiting the difference between the absorption and $\chi^{(3)}$ profiles for an appropriate two-photon-resonant intermediate level. Without phase matching and by using mJ lasers with a repetition rate of 10 Hz, we generate 0.23 μJ per pulse at 104.8 nm.

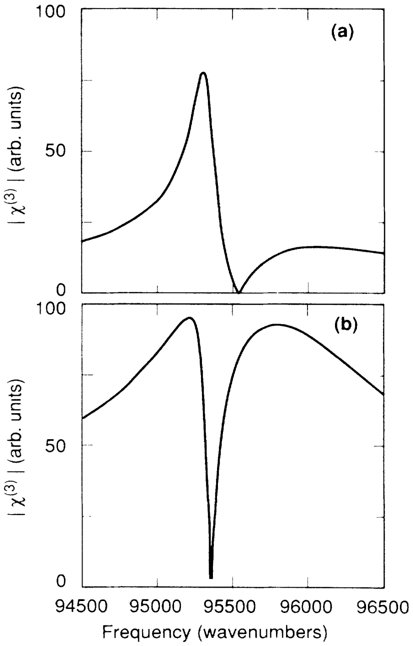

FIG. 4. Calculated nonlinear susceptibility with two-photon-resonant intermediate level (a) $3d^{10}4s4d\,^3D_2$ and (b) $3d^{10}4s4d\,^1D_2$.

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