Ionization-induced blue shift of KrF laser pulses in an underdense plasma

Y. M. Li* and R. Fedosejevs
Department of Electrical Engineering, University of Alberta, Edmonton, Canada T6G 2G7

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The ionization-induced blue-shifted spectra for helium, neon, and nitrogen have been measured at various gas densities up to 5×10^{20} cm^{-3} at a vacuum intensity of 8×10^{16} W/cm^2 for picosecond KrF laser pulses at 248 nm. A 1-mm diameter gas jet target was used in the experiment to minimize the refraction of the laser beam and thus higher laser intensities were obtained in the gas than in previously reported experiments. For helium, a distinct shifted peak was observed at intermediate densities which was not seen before. For helium and nitrogen, spectra were also measured of the light scattered outside of the original focal cone angle. In this region there was little signal for electron densities below 2×10^{20} cm^{-3} consistent with limited refraction at lower densities and at higher densities the spectra were predominantly blue shifted. These results indicate the importance of refraction in the correct interpretation of ionization blue-shifted spectra. [S1063-651X(96)12908-1]

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Spectral blue shifting from rapidly ionizing plasma was initially studied [1] using a nanosecond CO_2 laser incident on a static gas cell in the early 1970s, where the dominant ionization process was collisional ionization. Picosecond and femtosecond pulses have been used more recently [2–5] to carry out measurements of spectral blue shifting in rare gas targets in a totally new regime where the ionization mechanism is rapid optical-field-induced ionization resulting from high field intensity. Due to the extremely high ionization rate, there is a rapid creation of free electrons, causing a sudden decrease of the refractive index. Since the recombination time of the electrons in the plasma is much longer than the pulse width of the laser, the electron density increases monotonically throughout the laser pulse. The resultant negative time rate of change of the refractive index leads to blue-shifted components in the spectrum of the transmitted radiation. Theoretical modeling and computer simulations can be used to interpret the dynamics of the plasma ionization process [6,7] based on the resultant spectrum and, in principle, can be used to test various models of optical-field-induced ionization.

At the same time, the electron density gradients, and thus refractive index gradients, formed by the ionization process will also lead to defocussing of the main laser beam. This significantly reduces the peak intensity which can be achieved when focusing into a static gas cell as evidenced in previous numerical studies [8–10] and in the results to be presented below. The previous experimental studies of ionization blue shift [2–5] were all carried out in static gas cells for pressures up to 40 atm with vacuum intensities of 10^{13}–10^{16} W/cm^2. In order to match the theoretically calculated spectra with experimentally observed spectra peak intensities of 5–10 times lower than these vacuum focus intensities were required [5,6].

In this communication, the blue-shifted transmitted spectra of two rare gases (helium and neon) and a molecular gas (nitrogen) are presented for interaction of 1 ps duration, 248.5-nm KrF laser pulses at vacuum focal intensities of 8×10^{16} W/cm^2. One unique feature of the present study as compared to previous studies is that the laser pulse is focused onto a gas jet target [11] rather than into a static gas cell, so that refraction can be reduced and higher intensity can be achieved at high gas densities. At high gas densities the spectrum of the radiation refracted outside of the original beam cone angle was also measured for helium and nitrogen.

The initial picosecond KrF laser pulse is generated using a dye laser seed pulse which is frequency doubled and amplified in a double pass geometry through a discharge laser module. This pulse is then spatially filtered and amplified in an e-beam amplifier [12]. The 4×5 cm^2 cross-section pulse was focused using an aspheric doublet lens with a focal length of 20 cm, yielding a focal spot of 18 μm in diameter. For typical pulse energies of 200 mJ and pulse duration of 1 ps, the vacuum focal spot intensity was 8×10^{16} W/cm^2. The KrF laser radiation was focused at the center of the gas jet produced from a 1-mm diameter nozzle. The density of the gas jet was measured interferometrically [11] giving gas densities of up to 5×10^{20} cm^{-3} at a distance of 300 μm above the nozzle tip where the laser was focused. The transmitted and forward scattered KrF light was collected in a full cone angle of 25° using a UV triplet lens with a focal length of 12.5 cm. The gas jet was located in an evacuated target chamber which was pumped to a base pressure of 10^{-3} Torr before each shot. The light entered and exited the evacuated interaction chamber via CaF_2 windows to minimize any additional absorption. Part of the transmitted beam, reflected off of a beam splitter, impinged on a diffuser to average the radiation from the beam profile. An optical fiber lying behind the diffuser directed the radiation to the entrance of a 586-mm focal length Ebert monochromator which had a grating of 2880 lines/mm. The resolved spectrum was recorded using an optical multichannel analyzer (OMA) with an overall system resolution of 0.08 nm. Part of the input spectrum was sampled by a second fiber imaged through the spectrograph to a different spatial position on the OMA giving an input reference spectrum for every laser shot. When

*Present address: Columbia University, Dept. of Electrical Engineering, New York, NY 10027.

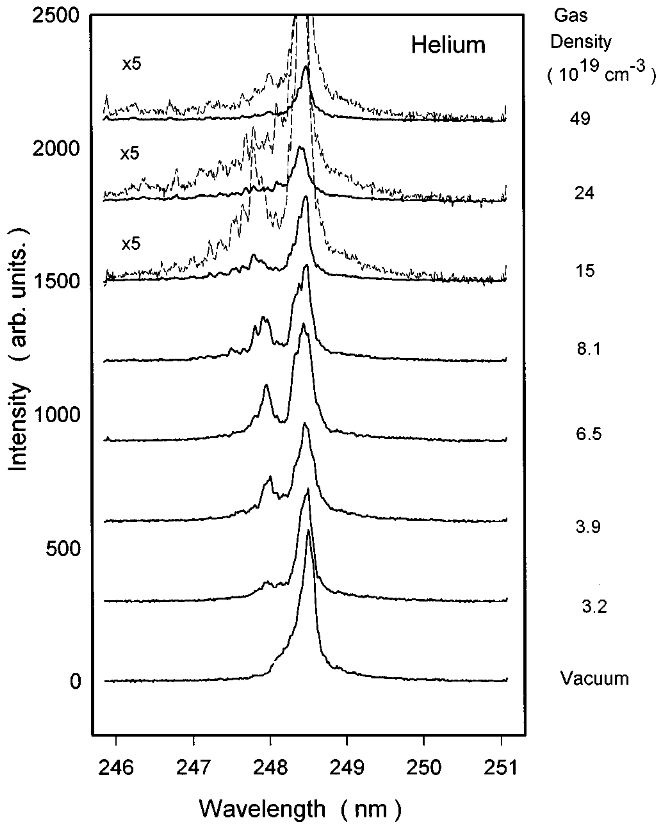

FIG. 1. Transmitted spectra for helium at various gas densities for a vacuum intensity of 8×10^{16} W/cm^2. The top three curves also show five times magnified curves of each.

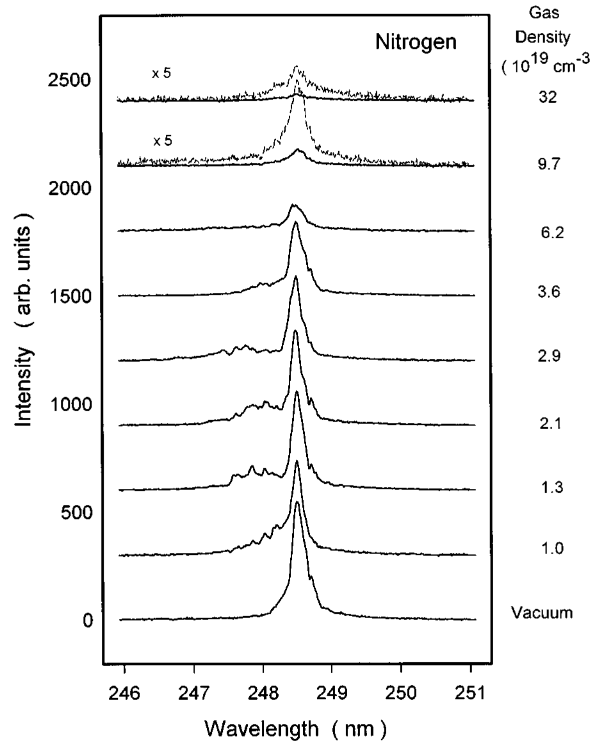

FIG. 2. Transmitted spectra for nitrogen at various molecular gas densities for a vacuum intensity of 8×10^{16} W/cm^2. The top two curves also show five times magnified curves of each.

no gas jet was employed, i.e., shots in vacuum, there was no measurable change in the output pulse spectrum caused by self-phase modulation or other effects along the path of the transmitted beam.

The dependence of the transmitted spectrum on gas density for helium gas at a vacuum laser intensity of 8×10^{16} W/cm^2 is shown in Fig. 1. The intensity scale is in arbitrary units but the relative number of counts is meaningful between the various spectra. It can be seen that at a low atomic density of 3.2×10^{19} cm^{-3} a slight shoulder emerges from the blue side of the pulse. As the density increases, a distinct peak is formed at densities of the order of 6×10^{19} cm^{-3}, which is blue shifted by 0.54 nm from the center of the input beam. This shifted peak presumably corresponds to the leading edge of the pulse in which the ionization process predominantly occurs. The remainder of the pulse which propagates through the already ionized plasma shows up as the main peak which is only slightly blue shifted by $\sim$0.06 nm. The height of this peak is reduced as expected since the energy is channeled into the blue-shifted spectral band. As the gas density is increased even further, the shifted peak starts breaking up into a long modulated shoulder extending towards shorter wavelength. As this occurs the radiation is spread over an even larger wavelength range and the observed spectral intensity decreases as seen in the figure.

The transmitted spectra for nitrogen and neon are shown in Figs. 2 and 3, respectively. The overall dependence of the blue-shifted spectra is similar to that for helium. However,

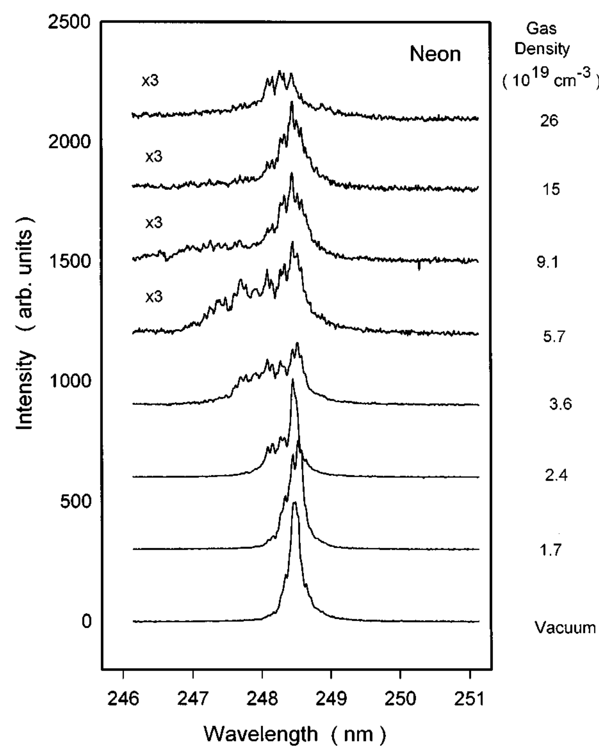

FIG. 3. Transmitted spectra for neon at various gas densities for a vacuum intensity of 8×10^{16} W/cm^2. The top four curves have been magnified three times in the vertical scale.

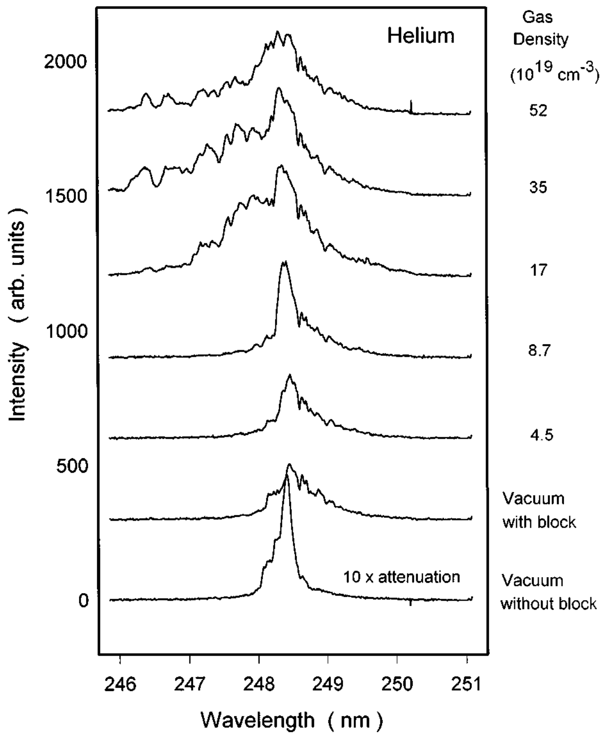

FIG. 4. Transmitted spectra with a beam stop for helium at various gas densities for a vacuum intensity of 8×10^{16} W/cm^2. The bottom curve is 10 times attenuated.

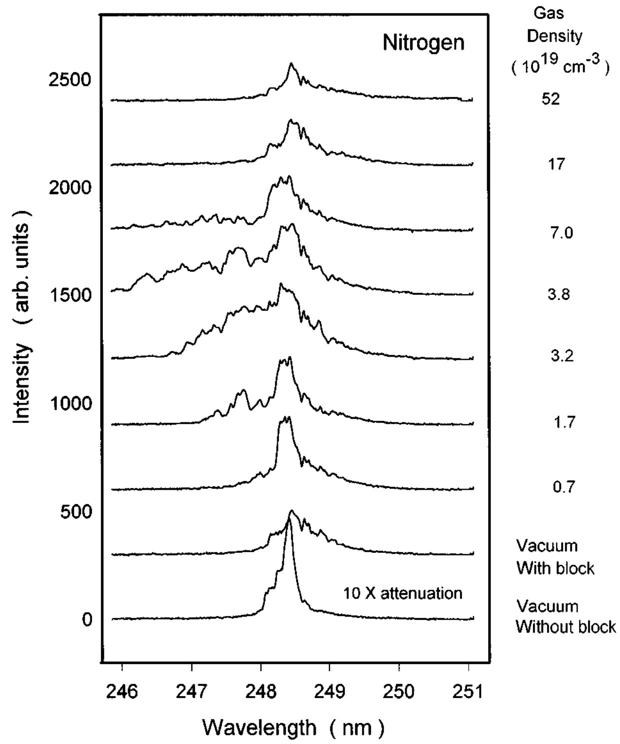

FIG. 5. Transmitted spectra with a beam stop for N_2 at various molecular gas densities for a vacuum intensity of 8×10^{16} W/cm^2. The bottom curve is 10 times attenuated.

there are some differences. First, as these are higher-Z species which are ionized to higher levels, much less gas density is required to produce the same density of electrons and give a comparable blue shift. For a peak intensity of 8×10^{16} W/cm^2 the expected ionization states of helium, nitrogen, and neon are 2, 5, and 6, respectively for a 1-ps pulse. Thus, 5 and 3 times lower gas densities would be required to achieve the same electron densities for nitrogen and neon, respectively (two atoms for each N_2 molecule). Second, while a distinct blue-shifted component is observed, no distinct gap is observed separating the blue-shifted part from the unshifted peak as for helium. Third, a significant amount of fine structure appears, which has not been seen in the previously reported investigations. Since none of the spectra shows a significant degree of red shifting we can conclude that the observed shifts result from ionization during plasma formation rather than the nonlinear response of the neutral medium, which would give a predominantly red shift [13].

To investigate the relation between refraction of the input pulse and the ionization induced blue shift, a stop, the same size as the unrefracted laser beam, was placed in the transmitted light diagnostic beam path such that the original beam path was totally blocked. Thus the light which could pass around the stop was that which had been refracted out of the original beam cone by the plasma. With this block in place, the measured transmitted spectra for helium are shown in Fig. 4 for different gas densities. The bottom curve is a vacuum shot without the stop in place but with 10 times attenuation. The next curve up is the scattered light signal with the block in place and no gas present. With gas densities up to 8.7×10^{19} cm^{-3}, there is predominantly scattered light around the original wavelength similar to the vacuum shot but no significant blue-shifted spectra. In comparison with the blue-shifted spectra obtained without the block in Fig. 1, this implies that the blueshifted component of the spectrum in this density range is mainly confined to the original beam path and thus is blocked by the stop. With increasing density, a strong blue-shifted spectral band appears as shown in the top three curves in Fig. 4. Note that the spectral component at the input wavelength does not increase significantly from its vacuum stray light level indicating that light which is strongly refracted is also predominantly blue shifted. This indicates that regions with large electron density gradients correspond to regions of rapid ionization and blue shift. As shown in Fig. 5, similar spectra were observed for nitrogen but with onset of the phenomena at much lower gas densities as expected from the higher degree of ionization. In this case, with densities higher than 3.8×10^{19} cm^{-3}, the blue-shifted spectra extends to shorter wavelengths, leading to weaker and weaker spectral intensity in the range of 246–248 nm as also observed without the spatial beam block.

A homogeneous model can be used to make a simple estimate of the blue shifts [6]. For a homogeneous plasma produced by monochromatic laser light of wavelength λ_0, under the assumption that $N_e \ll N_{cr}$, the spectral shift is given by

$$\Delta\lambda = -1.25\times10^{-13}\frac{e^2 N_i \lambda_0^3 L}{8\pi^2 m_e c^3}\frac{dZ}{dt}, \tag{1}$$

where N_i is the ion density, Z is the degree of ionization, and L is the interaction length. λ_0 and $\Delta\lambda$ are in units of nm and the rest of the constants have their usual meaning in cgs units. It is clear that the shift is very much dependent on the ionization rate. In the present experimental conditions for helium as in Fig. 1, if we take the case of density $N_i = 6.5\times 10^{19}$ cm^{-3} and an interaction length over which strong ionization of helium is expected, $L=0.4$ mm, we have $\Delta\lambda = 0.59-5.9$ nm assuming that $dZ/dt = 10^{12}$–10^{13} s^{-1}. The measured shift of the second peak is 0.54 nm to the blue. Thus this estimation gives the right order of magnitude of shift if the slower ionization rate is chosen or the interaction length is shorter than 0.4 mm.

More accurate analysis requires numerical modeling of the beam propagation and ionization process. Such modeling of the ionization-induced blue shift has been carried out using one-dimensional [6,7] and two-dimensional [5,10] electromagnetic propagation codes. The one-dimensional models could only qualitatively describe the observed spectra and only gave agreement when intensities approximately 10 times lower than the experimental intensities were employed. The comparison of the two-dimensional model, which included refraction with experimental data, gave better quantitative agreement and predicted the reduction in intensity due to defocusing upon the plasma formation [5,10]. These simulations revealed different spectral shifts for different radial positions across the beam with the largest shifts occurring at the edges of the beam for high intensities and high densities. Also, they predicted increasing spectral structure for increasing intensities and gas pressures. Qualitatively our results agree with these predictions. We see significant refraction at higher gas densities as evidenced by the radiation transmitted around the beam block. The outside of the beam, which is transmitted around the beam block, is predominantly blue shifted. Also, we observe significant structure in the transmitted spectra as evidenced by the distinct peak for helium gas and detailed structure for the higher-Z gases. This presumably arises from the use of a gas jet target allowing higher intensities to be achieved at higher interaction densities. Ideally such modeling calculations should be carried out for our exact experimental conditions and detailed comparisons made.

In summary, the blue-shifted spectra for helium, neon, and nitrogen have been measured at various pressures at a vacuum intensity of 8×10^{16} W/cm^2 when irradiated with picosecond KrF laser radiation. A gas jet target was used in this experiment to reduce the refraction of the laser beam and thus higher laser intensities could be obtained at high gas densities. For helium, a distinct second peak was revealed and was attributed in part to the high intensities achieved in the present gas jet target. At the lower densities, the blue-shifted spectra were confined mainly to the original beam path. However, as the pressure increased, the laser beam was refracted and the refracted beam also exhibited a predominantly blue-shifted component. This work confirms the expectation of strong blue shifting occurring for the strongly refracted component of the light.

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