Application of cold beam of atoms and molecules for studying luminescence of oxygen atoms stimulated by metastable helium

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Abstract. We describe a method for creating a high flux beam of cold atoms and molecules. By using this method, spectroscopic studies of the afterglow of oxygen-helium gas mixtures at cryogenic temperatures were performed. The cooling by helium vapor of a helium jet containing trace amounts of oxygen after passing through a radiofrequency discharge zone led to the observation of strong emissions from atomic oxygen. The effect results from the increased efficiency of energy transfer from metastable helium atoms and molecules to the atomic oxygen in the cold dense helium vapor. The effect might find application for the detection of small quantities of impurities in helium gas as well as possible laser action.

1. Introduction
The studies of inelastic collisions of cold atoms and molecules have recently attracted theoretical and experimental interest [1, 2]. Cold molecules are ideal objects for precise measurements of fundamental constants [3, 4]. Both these areas of research benefit from high flux sources of cold atoms and molecules. Recently a large variety of techniques to produce cold, low velocity samples of atoms and molecules were developed [5, 6, 7]. The brightest source of cold molecules \((3 \times 10^{12} \text{ s}^{-1})\) was based on the buffer gas cooling method [8].

In this work we present a method for obtaining cold atoms and molecules that uses a hydrodynamic effect to enhance the beam flux by two orders of magnitude as compared to the best existing sources. By using this method we studied processes of energy transfer from metastable He atoms and molecules to oxygen atoms. This method is based on the injection of a gaseous jet containing a small admixture of impurities which passed through a radio frequency discharge into superfluid helium [9]. When the impurity content in the helium gas is of order 1%, this method is very efficient for producing nanoclusters [10]. In the present work we found that reducing the ratio between impurity content and helium atoms in the jet from \(10^{-2}\) to \(10^{-5}\) allows one to avoid the impurity clustering process in the cold helium gas and to create a beam of cold atoms and molecules. As the jet descends into superfluid helium (see Fig. 1) the helium and impurity atoms and molecules in the jet are cooled efficiently by the dense helium
vapor and the temperature of the atoms in the jet is decreased from 150 K near the orifice to 1.4 K near the surface of HeII. The process of cooling occurs over a distance of \(~2.5\) cm. The average travel time of an atom or molecule from the orifice to the surface of superfluid helium is of order 100 \(\mu s\). Although we cannot obtain direct spectroscopic evidence for the temperature of the oxygen atoms, following Ref.[5], we can estimate the collision numbers needed for ”warm” (\(~150\) K) oxygen and helium atoms injected from discharge zone to be fully thermalized to a temperature of 1.4 K. These numbers are \(~30\) collisions for oxygen atoms and \(~20\) collisions for helium atoms. Near the surface of HeII in the cold dense helium gas, oxygen atoms undergo 300 collisions in 1 \(\mu s\) [11], which leads to full thermalization of the oxygen and helium atoms in the jet to 1.4 K. The temperature of excited atoms and molecules produced by energy transfer from metastable helium in the lower part of the jet might be higher than the temperature of thermalized atoms and molecules.

**Figure 1.** (Color online) Scheme of the set-up for creation of a high flux beam of cold atoms and molecules and the spectroscopic study of the afterglow in cold helium vapor. 1-helium dewar, 2-quartz tube, 3-space filled with liquid nitrogen, 4-electrodes for radiofrequency discharge, 5-system for registration of optical spectra, 6-gas jet, 7-lens, 8-quartz beaker, 9-superfluid helium in the beaker, 10-level of liquid helium in the main helium bath, 11-fountain pump.

2. Experimental setup
The overall experimental arrangement is shown in Fig. 1. The gas mixture consisting of helium and small concentrations of impurities enters the helium bath region via a quartz capillary cooled by liquid nitrogen. Electrodes placed around the quartz capillary provide a 53 MHz, 90 Watt radiofrequency discharge to form metastable helium atoms and dissociate the oxygen molecules. In the experiment, we used research grade helium from Linde Electronics & Specialty Gases with 99.9999% purity. The oxygen content in the discharge space (1 ppm) results from contamination of the helium gas. The content of impurities in the gas mixture can be adjusted by adding trace amounts of impurities to the helium gas or by purifying the helium gas in a cold trap at liquid helium temperatures. Under a small gradient of pressure (\(\Delta p \sim 2\) Torr) between the discharge zone and the cryostat, a well-formed jet is created after passage of the gas mixture through a small orifice (0.75 mm dia) in the quartz capillary. The flux of \(5 \cdot 10^{19}\) atoms/s of the impurity-helium mixture entering the cryostat is measured by a Brooks Instrument 5850E flow controller. A temperature of 1.4 K of the liquid helium in the cryostat is maintained by pumping the main helium bath and was measured by a germanium thermometer. A collection beaker was placed below the orifice with a distance of 2.5 cm between the orifice and the top of the beaker. The spectra of the gas jet afterglow were studied when the beaker was empty and also when the beaker was filled with superfluid helium by the fountain pump shown in Fig. 1. We studied the dependence of spectra on the pressure of helium gas in the helium dewar by varying the pumping speed on the main bath. We also studied the dependence of spectra on
oxygen content in the oxygen-helium gas mixture. In these experiments the light from the jet afterglow was collected near the top of the collection beaker at a distance of 2.5 cm below the orifice by a lens which focused it onto the end of an optical fiber (see 7 on Fig. 1). The other end of the fiber was attached to an Andor Shamrock SR500 spectrograph. The Andor spectrograph with a Newton EMCCD camera was used to obtain high resolution (0.05 nm) spectra over a broad wavelength range (200 -1100 nm).

![Figure 2](image)

**Figure 2.** (Color online) Spectra of the helium jet with traces of oxygen (1 ppm): a) when the beaker is filled with HeII and b) when the beaker is empty. Light is collected at the top of the beaker c) High resolution spectra of the atomic oxygen lines. The spectra with larger intensity are from the jet when the beaker is filled with HeII while the ones with smaller intensity are from the jet when the beaker is empty. d) High resolution spectrum of the He2 band near 639.6 nm. The He2 band from cold jet was shifted up vertically for better comparison.

3. Experimental results

In the method we used, the impurities and helium gas in the jet are moving in the same direction in the cold dense helium gas. During the cooling process, the velocities of atoms in the jet are reduced substantially. Relative velocities between helium atoms and impurities become negligible, and it is possible to study processes of energy transfer and chemical reactions in the gas phase at low temperatures. As an example, in this work we have studied the influence of temperature on the spectra of oxygen atoms excited by metastable helium atoms and molecules [12]. Fig. 2a and 2b show the emission spectra of helium jet with 1 ppm concentration of
oxygen detected by the Andor spectrometer for the cases when the collection beaker contained liquid helium and when it was empty. Both spectra contained atomic oxygen lines as well as atomic helium lines and molecular helium bands listed in Table 1. When the beaker was empty, helium atomic lines and molecular bands dominate the spectrum. Only three weak oxygen lines were present in this spectrum (see Fig. 2b). When liquid helium was filled into the beaker, the oxygen lines become much stronger (see Fig. 2a and 2c) whereas the He\(_2\) molecular bands became smaller. High resolution spectra for the oxygen lines and He\(_2\) molecular band are shown in Fig. 2c and 2d. The fine structure obtained helped to definitely identify the spectral lines observed. The unresolved spectra of oxygen atomic lines were obtained earlier in studying helium jets with unknown contaminations of oxygen in similar conditions, but the authors did not make identifications of oxygen lines in their work [13]. In our experiment we observed a large enhancement of the oxygen line intensities during filling the collection beaker with superfluid helium, which provided conditions for efficient cooling of the oxygen-helium jet.

We studied experimental conditions for which the large enhancement of the oxygen line intensities could be observed. The spectra shown in Fig. 2 were obtained at the helium vapor pressure in the cryostat of 2 torr and for the ratio between O\(_2\) and He in the gas jet equal to 10\(^{-5}\). Fig. 3a shows the dependence of intensities of atomic lines and molecular bands on the pressure of helium vapor in the cryostat. It was found that as the pressure increased up to 12 torr, the intensity of all spectral lines and bands gradually decreased. Fig. 3b shows the dependence of the intensities of atomic oxygen and helium lines and He\(_2\) molecular bands on the content of O\(_2\) molecules in the oxygen-helium gas mixture. The dependence of intensities of different oxygen atomic lines have maxima in the range 20-200 ppm of O\(_2\) gas mixture. The intensity of He atomic line does not change upon changing concentration of O\(_2\) molecules. However, the intensity of the He\(_2\) molecular band decreases when the content of O\(_2\) molecules in the gas mixture increases. This indicates that the observed bright green luminescence of O atoms requires a very special set of conditions. For pressures above 12 torr and temperatures above 1.8 K the phenomena was not observed. At oxygen contents above 400 ppm in helium gas, the effect also became very weak.

4. Discussion
In our experiments the average travel time of an atom or a molecule from the orifice to the surface of superfluid helium is of order 100 \(\mu\)s. This means that all atoms and molecules excited in the

### Table 1. Identification of observed lines and bands shown in Fig. 2.

| \(\lambda\), nm | Transition | \(\lambda\), nm | Transition |
|----------------|------------|----------------|------------|
| 844.6          | O I (3s\(^3\)S-3p\(^3\)P) | 477.3          | O I (3p\(^3\)P-7d\(^3\)D) |
| 777.4          | O I (3s\(^5\)S-3p\(^5\)P) | 728.3          | He I (2p\(^1\)P-3s\(^1\)S) |
| 645.4          | O I (3p\(^3\)P-5s\(^5\)S) | 706.5          | He I (2p\(^3\)P-3s\(^3\)S) |
| 615.7          | O I (3p\(^3\)P-4d\(^5\)D) | 667.9          | He I (2p\(^1\)P-3d\(^1\)D) |
| 604.6          | O I (3p\(^3\)P-6s\(^3\)S) | 587.6          | He I (2p\(^3\)P-3d\(^3\)D) |
| 555.6          | O I (3p\(^3\)P-7s\(^5\)S) | 501.6          | He I (2s\(^1\)S-3p\(^1\)P) |
| 543.6          | O I (3p\(^3\)P-6s\(^5\)S) | 639.6          | He\(_2\) (d\(^3\)\Sigma^+ \rightarrow b\(^3\)\Pi_g) |
| 533.0          | O I (3p\(^3\)P-5d\(^5\)D) | 573.2          | He\(_2\) (f\(^3\)\Delta_u \rightarrow b\(^3\)\Pi_g) |
| 502.0          | O I (3p\(^3\)P-7s\(^5\)S) | 465.0          | He\(_2\) (e\(^3\)\Pi_g \rightarrow a\(^3\)\Sigma_u^-) |
| 496.8          | O I (3p\(^3\)P-6d\(^5\)D) | 454.7          | He\(_2\) (h\(^3\)\Sigma_u^+ \rightarrow b\(^3\)\Pi_g) |
| 480.2          | O I (3p\(^3\)P-8s\(^5\)S) | 445.0          | He\(_2\) (i\(^3\)\Sigma_u^- \rightarrow b\(^3\)\Pi_g) |
discharge zone with lifetimes in the range 14 - 1000 ns should emit light only in the discharge zone and near the orifice. Therefore the observed emission spectra well below the orifice should result from processes taking place in the jet. It is well known that metastable helium species easily excite, dissociate, and ionize any admixtures of impurity species in helium plasmas and in the afterglow [14, 15, 16]. The process of Penning ionization (PI) of molecular oxygen by helium atoms and molecules has been intensively studied [17]. In our experimental condition when we observed bright green emission, the very low content (1-25 ppm) of O$_2$ molecules in helium gas are completely dissociated in the discharge zone; therefore all of the oxygen should be in an atomic state in the expanded helium jet. The PI processes of excitation of oxygen atoms by helium atoms and molecules is a dominant process in the jet. Formation of highly excited states of oxygen atoms is a result of PI of oxygen atoms by helium atoms and molecules followed by electron-oxygen ion recombination. Qualitatively the effect of large enhancements of the oxygen line intensities and the appearance of new, even more intense oxygen lines in the dense cold helium vapor observed in this work might be explained by an increase in density of the cold oxygen and metastable helium atoms and molecules as the jet enters the region of dense helium vapor near the surface of liquid helium. The process of the PI of O atoms should be much more efficient in this region. Another possible factor for enhancement of the oxygen line intensities is the increasing cross sections of PI processes for oxygen atoms by metastable helium atoms and molecules at low temperatures. In our experiments the temperature of the atoms and molecules in the jet before entering bulk superfluid helium should be thermalized to the temperature close to the temperature of superfluid helium and both reacting species, helium and oxygen, are moving in the same direction, representing a situation with cold merged beams, which provides an increasing time for interaction of the reagents [18, 19]. An example of the consequences of these increased interaction times of the slow beams at low temperatures was the observation of quantum effects during the PI process [19].
The bright green triplet lines of 543.6 nm (see Fig. 2c) are emitted from the 6s\(^5\)S state which has a long decay time (0.259\(\mu\)s) whereas the final state lifetimes corresponding to these transitions are much shorter (\(\sim 27\) ns). This provides the possibility for developing a population inversion between the 6s\(^5\)S and 3p\(^5\)P levels to study the lasing at \(\lambda=543.6\) nm of atomic oxygen. An energy level diagram and the transitions of the oxygen atom involved in the lasing process are shown in Fig. 4. An atomic oxygen laser was first operated with an RF discharge in Ne-O\(_2\) and Ar-O\(_2\) mixtures at \(\lambda=844.67\) nm [20]. The excitation mechanism involving energy transfer collisions between metastable rare gas atoms and ground state O\(_2\) molecules to generate a population inversion was analyzed later [21]. We plan to investigate the possibility of laser action at \(\lambda=543.6\) nm in a cold oxygen-helium jet excited in a RF discharge.

**Conclusion**

All spectral lines and bands observed in this work correspond to oxygen and helium atoms and helium molecules in the gas phase. We did not observe any sign of recombination or clusterization of oxygen atoms. This means that we had realized conditions for the creation of an intense cold gas phase oxygen-helium beam in which the flux of oxygen atoms is of order \(3 \cdot 10^{14}\) atoms/s. This flux is almost two orders of magnitude larger than that obtained by using a buffer helium gas method [5, 8]. In our experiments this flux is injected into the volume of superfluid helium, but it is possible to realize a geometry in which the jet will pass the region of cold helium gas near the surface of HeII and enter a high vacuum region for further investigations and applications.

The effect of substantial enhancement of the intensity of atomic oxygen luminescence by the cooling of a helium jet with small traces of O atoms by helium vapor was observed. The effect was explained by effective energy transfer from metastable helium atoms and molecules to oxygen atoms and molecules under cryogenic conditions. This effect might provide a practical method for the detection of small traces of impurities in helium.

Observation of the intense atomic oxygen emission at \(\lambda=543.6\) nm in cold oxygen-helium jets after RF discharge opens possibility of creating population inversions of atomic oxygen. We plan experiments to search for laser action of atomic oxygen at \(\lambda=543.6\) nm in the cold oxygen-helium beam excited by an RF discharge.
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