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Laser spectroscopic study of Mg atoms in pressurized liquid helium

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Abstract

Emission and excitation spectra of the $3s^2\ ^1S_0$–$3s3p\ ^1P_1$ transition of Mg atom in pressurized liquid helium-4 and helium-3 have been measured. In the emission spectra we have found that their transition wavelengths, which are significantly red shifted, remain constant or slightly increase with increasing the liquid helium pressure, whereas for other alkali–earth atoms the increase of the liquid pressure always shifts their emission spectra toward shorter wavelength. Our theoretical calculations based on a bubble model have successfully reproduced this unique spectral property, and have suggested the possibility of the formation of a $Mg(3s3p\ ^1P_1)He_n$ exciplex in a bubble.

Keywords: Laser spectroscopy; Mg atom; Liquid helium; Bubble

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1. Introduction

In the past two decades, a number of atomic species implanted in liquid helium have been studied with spectroscopic methods [1,2]. These studies have so far revealed not only structural properties of helium atoms surrounding impurity atoms, such as bubble-like cavity structure and exciplex formation, but also physical properties of liquid helium itself, such as superfluidity and roton–maxon excitation [1].

Liquid helium is a unique substance, which remains liquid even at the absolute zero and solidifies only at a high pressure no less than 2.6 and 5 MPa for ^{4}He and ^{3}He, respectively. This means that liquid helium is quite compressible, and that even a small change of liquid pressure causes significant variations in the number density of helium atoms and in the interatomic distance between the impurity and its surrounding He atoms. For this reason, it has been attracting our interest to spectroscopically study impurity atoms as a function of the liquid pressure, and many authors have so far reported such spectroscopic investigations.

Kanorsky et al. have studied Ba atoms implanted in pressurized liquid helium and solid helium [3]. They have shown that in both excitation and emission spectra of the $6s^1S_0$–$6p^1P_1$ transition their transition wavelengths, which are blue shifted, decrease with increasing the pressure, and that this can be explained by the bubble model. Kinoshita et al. have studied Rb and Cs atoms in pressurized liquid helium, and have found similar pressure dependences of their spectral properties [4]. They have also shown that the D1 emission of Rb is quenched as the pressure increases [5], and that this quenching is attributed to the tunneling of He atoms through a small barrier of the $Rb(5p^2\Pi_{1/2})$–He pair potential. This leads to the formation of exciplex and then their dissociation succeeds, which is a non-radiative decay process of the $5p^2P_{1/2}$ state of Rb. As for an exciplex of Cs, Nettels et al. have recently discovered a novel exciplex of $Cs(A\Pi_{1/2})He_7$ in the hpc phase of solid helium [6]. Similar studies on the pressure dependence have also been carried out for the transition between the $4f^{13}(^2F_0^{7/2})6s^2$ state and a mixed state of the $4f^{12}(^3H_6)5d_{5/2}6s^2(6,5/2)_{7/12}$ and $4f^{13}(^2F_0^{7/2})6s6p(^3P_0^0)(7/2,0)_{7/2}$ states of Tm [7], and for the 3s4 s $^3S_1 \rightarrow 3s3p\ ^3P_J$ ($J=0$, 1 and 2) transitions of Mg [8]. These studies have shown that the pressure dependence of the spectral shifts are similar to those of Ba, Rb and Cs, while extremely narrow spectra have been observed for inner-core transitions of Tm atoms.

In the present study, we have studied the $3s^2\ ^1S_0$–$3s3p\ ^1P_1$ transition of Mg atoms in pressurized liquid helium. Emission and excitation (absorption) spectra have been measured for liquid helium at the saturated pressure for 1.4 K. In particular, emission spectra of this transition are considerably broadened

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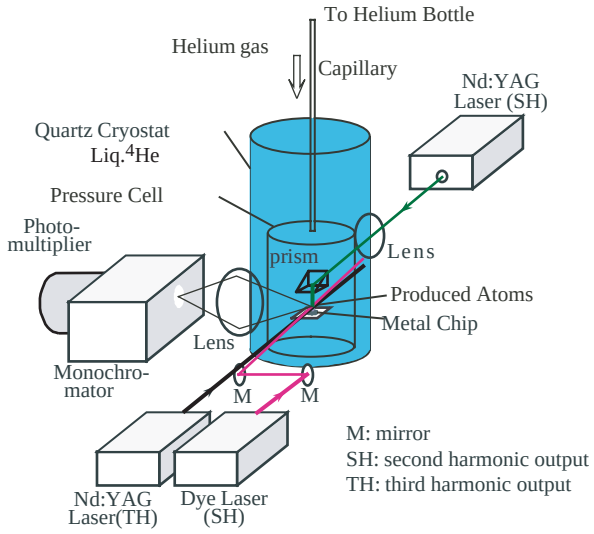

Fig. 1. Experimental setup.

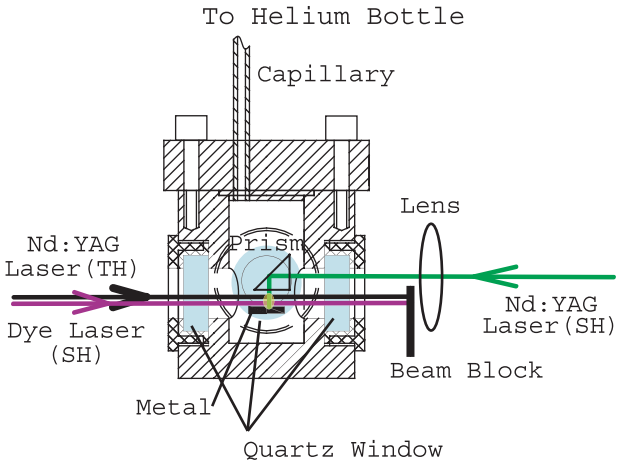

Fig. 2. Detailed configuration of the pressure cell.

in comparison with those of other alkali–earth atoms, so that it is interesting to study their dependence on the liquid pressure. Similar measurements have also been carried out for pressurized liquid ^{3}He.

2. Experimental

The experimental setup is shown in Fig. 1. Helium-4 gas (99.9999% in purity) or helium-3 gas (99.95%) is introduced into a pressure cell made of stainless steel through a capillary tube. The pressure cell is immersed in liquid ^{4}He kept at a temperature of 1.4 K, and the helium gas is gradually liquefied in the cell. The pressure of the cell is controlled by a pressurized gas cylinder with a regulator connected to the other end of the capillary tube, and is monitored by a semiconductor pressure gauge. Although the gas cylinder is capable of applying more than 6 MPa, a block in the capillary restricts the effective pressure in the cell below the solidification pressure, which is 2.6 MPa for ^{4}He and 5 MPa for ^{3}He.

Magnesium atoms are implanted into the liquid helium in the pressure cell by laser sputtering of a Mg metal chip placed in the liquid helium with two successive light pulses, which are second and third harmonic pulses of Nd^{3+}:YAG lasers, respectively; the width and energy of each pulse are 5 ns and 2 mJ, respectively (see Fig. 2). The Mg atoms thus produced are then excited by a second harmonic pulse of a dye laser (Fluorescane 548). Laser induced fluorescence (LIF) from the atoms is introduced into a 25 cm monochromator through a lens and detected by a photo-multiplier.

The transition studied is the one between the ground state ($3s^2\ ^1S_0$) and the first excited singlet state ($3s3p\ ^1P_0$). Its excitation spectra are measured by scanning the dye laser wavelength with the monochromator wavelength fixed at each emission peak, while emission spectra are measured by scanning the monochromator wavelength with the laser wavelength fixed at each absorption peak.

The excitation spectra observed for Mg atoms in liquid ^{4}He are shown in Fig. 3(a), and the emission spectra are shown in Fig. 3(b). As seen in Fig. 3(a), all the excitation spectra show broad widths and large blue shifts with respect to the transition wavelength in free space. With increasing the pressure of the liquid, their peak wavelengths decrease, while the widths increase. These are usual spectral characteristics for impurity atoms in liquid helium [8]. On the other hand, as for the emission spectra, they show broad widths and large red shifts, which are also usual for impurity atoms in liquid helium. However, their dependences on the liquid pressure are significantly different not only from those of the excitation spectra but also from those seen for other atoms implanted in liquid helium; that is, both the peak wavelength and spectral width are almost constant or slightly increase with increasing the liquid pressure. These spectral properties are shown in Fig. 4(a) and (b).

We have also measured the same spectra for pressurized liquid ^{3}He. Their spectral properties are shown in Fig. 4(c) and (d). Although each of the excitation and emission spectra shows roughly the same characteristics as the one for ^{4}He, we can see clear isotopic differences between liquid ^{3}He and ^{4}He; the spectral widths for liquid ^{3}He are considerably narrower than those for liquid ^{4}He, and the peak shifts for ^{3}He are smaller than those for ^{4}He. All these pressure dependences of

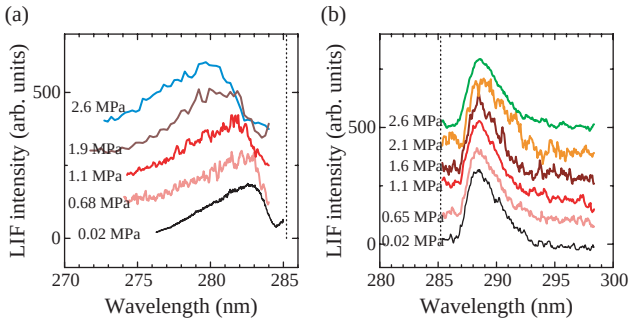

Fig. 3. Spectra observed for the $3s^2\ ^1S_0$–$3s3p\ ^1P_1$ transition of Mg atoms in pressurized liquid ^{4}He: (a) excitation spectra, (b) emission spectra. The vertical dotted line shown in each figure indicates the transition wavelength of Mg atoms in free space.