Implantation of Neutral Atoms into Liquid Helium by Laser Sputtering

A. Fujisaki, K. Sano, T. Kinoshita, Y. Takahashi, and T. Yabuzaki
Department of Physics, Faculty of Science, Kyoto University, Kyoto 606, Japan
(Received 3 May 1993)

We have developed a new method for producing a high density of neutral atoms in liquid helium; laser sputtering within liquid helium. When we irradiate samples immersed in liquid helium with a pulsed laser, a large number of clusters are introduced into the liquid. When we further irradiate these clusters with an additional pulse laser, the clusters are sputtered and decomposed, which results in the creation of a high density of isolated atoms in the liquid. For alkaline earth atoms this method is demonstrated, and the results of laser spectroscopy are given.

PACS numbers: 67.40.Yv, 79.20.Ds

Study of positive (He^+) and negative (e^-) ions in liquid and solid helium has revealed fundamental properties of this unique quantum liquid and solid [1]. Not only the motions of these charged impurities under electric fields but the structures have also given important informations on the surrounding helium [2]. Furthermore, the study of interactions between charged impurities and vortex lines and rings has been a quite important field of superfluid helium [3].

It is quite natural to expect that the study of neutral atoms in liquid and solid helium will also offer useful information for further understanding of these quantum systems. With optical and laser-spectroscopic techniques sensitive detection can be performed, and ultrafast dynamics can be investigated. Recently there has been an increasing interest in the system of atomic impurities in liquid helium [4] for atomic and elementary particle physics. Until quite recently there had been no method for creating various kinds of neutral atoms in liquid or solid helium [5]. There has appeared, however, a recombination method [6], with which alkaline earth atoms have been successfully implanted and the laser spectroscopy has been done [7].

In this paper we report on our newly developed method for implanting a high density of atoms into liquid helium; laser sputtering within liquid helium [8]. Many kinds of atoms can be rather directly created within liquid helium with this method. A detailed description of this method is given. Compared with the recombination method, our method is quite simple and has the unique feature of its applicability to pressurized liquid or solid helium. Furthermore, we have recently succeeded in detecting the signals of alkali atoms (Rb and Cs) in liquid helium for the first time with this method. In the present paper, for alkaline earth atoms (Ba and Ca) our laser sputtering method is demonstrated, and the laser-spectroscopic results are given, including those for newly found transitions of Ba and Ca in liquid helium.

Figure 1 shows an experimental setup of our new method for producing a high density of atoms in liquid helium. The experiment was performed at about 1.6 K and at the saturated vapor pressure. First we irradiated a sample (usually bulk metal samples of Ba or Ca [9]) immersed in liquid helium with a pulsed laser [10]. The laser beam was focused with a 5 cm lens onto the sample. At this stage one could see that direct products of this sputtering were mostly relatively large clusters or fine particles. When we further irradiated these clusters with a secondary pulsed laser beam, then it was the clusters that were sputtered and decomposed, which resulted in the creation of a high density of isolated atoms in the liquid. We could determine the existence of isolated atoms by exciting the atoms with a dye laser and detecting the emitted photons [11]. The probe dye laser traversed the same path as that for the secondary laser beam. Without the secondary laser beam the signals for atoms in liquid helium were absent. The photon counting

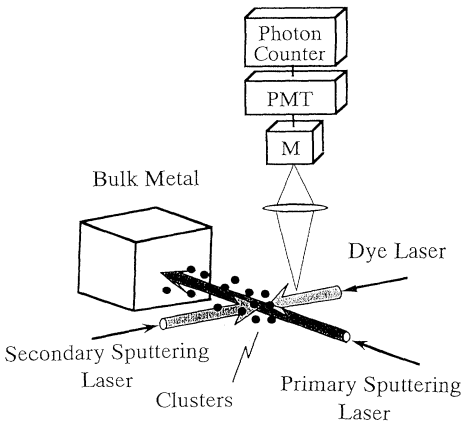

FIG. 1. The laser sputtering method to produce neutral atoms in liquid helium and the experimental setup for spectroscopy are shown. The clusters were produced by a primary sputtering laser, and a secondary sputtering laser decomposed the clusters into isolated atoms in liquid helium. The atoms in liquid helium were probed by a dye laser which passed through the same path as that of the secondary sputtering laser. The emitted photons were detected by a cooled photomultiplier tube (PMT) after a monochromator (M). The time for the photon counting was delayed by more than 5 μsec from the time of sputtering in order to avoid counting the scattered and recombination light as well as the emission at the instant of the secondary pulse.

0031-9007/93/71(7)/1039(4)$06.00

rate was normally as high as 10^5 counts per second with several hundreds of milliwatts of dye laser power. In some cases the emitted light could be observed with the naked eye. From the counting rate the atomic density is estimated to be about 10^8–10^{10} per cm^3 [12].

More simply one can successfully use a pulsed laser at a high repetition rate (about 1 kHz). The clusters produced by one laser pulse can be sputtered when the next pulse comes during the lifetime of the clusters in the liquid. In this case each laser pulse is effective for sputtering both the bulk sample and the clusters. In Fig. 2 we display the time-of-flight spectrum of the clusters in liquid helium produced by the primary laser sputtering. It shows that the clusters arrived at the observation region (separated by ~2 cm from the sample) about 1–2 msec after the primary sputtering. The clusters should have a broad velocity distribution at the moment of the primary sputtering, and convection or shock wave might be responsible for the observed bunching of the clusters. In the experiments the signals were most efficiently observed at about a 1 kHz repetition rate of the laser pulse and much less at lower rates, which is consistent with the result of Fig. 2. The signals shown in Figs. 3 and 4 were observed mainly with this method.

One might wonder why the sample can be heated and vaporized in superfluid helium which is so cold and has very high thermal conductivity with no drastic change of liquid temperature [13]. The region near the focal point (about 5 mm^3) consisted of helium vapor, as observed visually. It can be inferred that this region served as a thermal insulation from the superfluid bath [14]. Owing to this thermal insulator, the energy introduced by the pulsed laser did not dissipate completely into the bulk of superfluid helium, and was efficiently used for heating and vaporization of the sample. It is noted, on the other hand, that this sputtering method did not work in the normal state of liquid helium because the sputtering laser beam was scattered by bubbles produced around the sample.

One could observe strong emission of light from the path of the secondary laser beam at the moment of sputtering. The wavelengths of the emitted light were

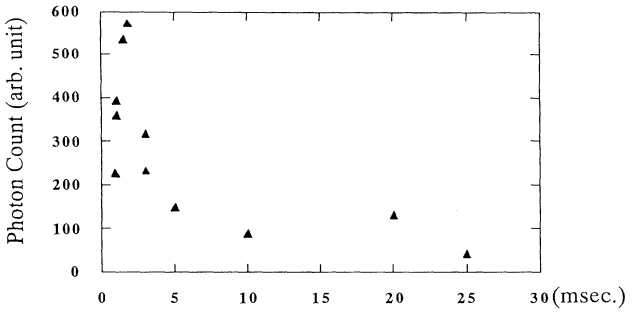

FIG. 2. Time-of-flight spectrum of the Ba clusters produced by laser sputtering in liquid helium. The intensities of the emission (near 553 nm) at the moment of the secondary laser sputtering are plotted against the delay time between the primary sputtering and the secondary one. The observation region is separated by about 2 cm from the sample. One can know that the clusters came to the observation region about 1–2 msec after the primary sputtering. The clusters should have a broad velocity distribution at the moment of the primary sputtering, and convection might be responsible for the observed bunching of the clusters.

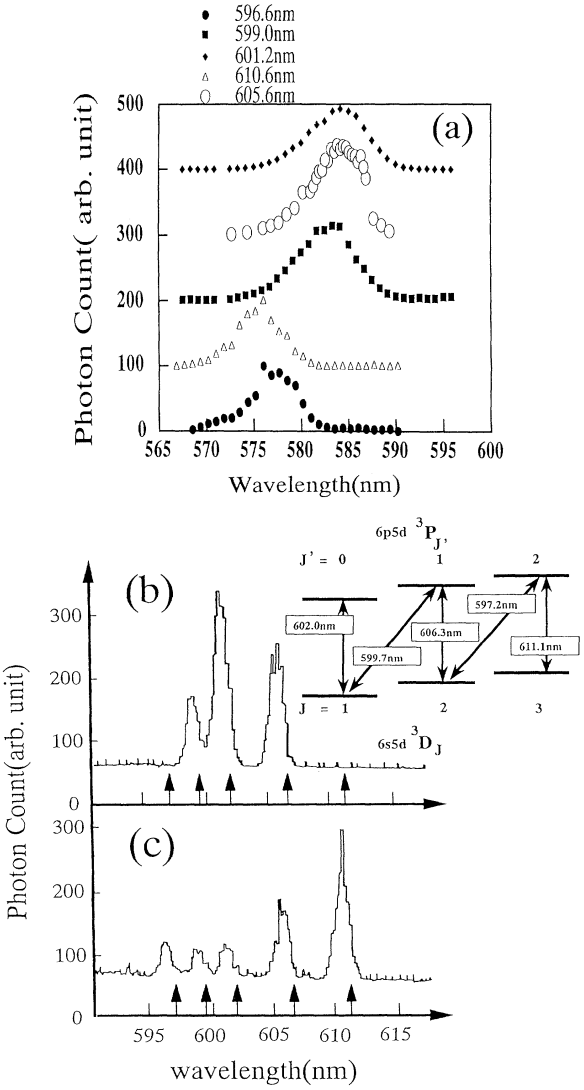

FIG. 3. Excitation and emission spectra of the triplet transitions of Ba. Energy level diagram of a free atom is shown in the inset. The excitation spectra (a) were obtained for each of the emission lines. The wavelength of the monochromator, which is set to that of each emission line, is indicated in the upper part of (a). The emission spectra (b) and (c) were obtained for the excitation at (b) 584 nm and (c) 574 nm, respectively. The resolution was about 0.2 nm. The arrows in (b) and (c) represent the positions of corresponding spectra of the Ba atoms in vacuum.

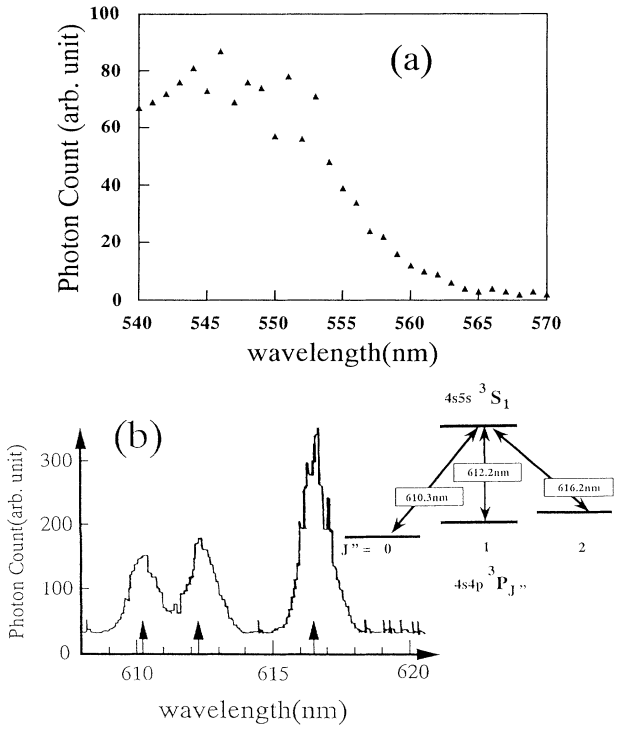

FIG. 4. Excitation (a) and emission (b) spectra of the triplet transitions of Ca. Energy level diagram of a free atom is shown in the inset. The excitation spectrum (a) was obtained for each emission line and was found to be almost the same for each. The data below 540 nm were not obtained because of the dye laser used (only R110 and R6G were used). The emission spectrum (b) was obtained for the excitation at 547 nm. The resolution was about 0.2 nm. The arrows in (b) represent the positions of corresponding spectra of the Ca atoms in vacuum.

near the free atomic transitions, and the spectra were as broad as about 1 nm. The transitions of $6s6s\,^1S_0 \leftrightarrow 6s6p\,^1P_1$ and $5d6s\,^3D_J \leftrightarrow 5d6p\,^3P_{J'}$ were observed for Ba, and $4s4s\,^1S_0 \leftrightarrow 4s4p\,^1P_1$ and $4s4p\,^3P_{J''} \leftrightarrow 4s5s\,^3S_1$ for Ca(J,J', and $J''=1$, 2, and 3, respectively). These are considered to be spontaneous emissions from isolated atoms in liquid helium initially created in the excited states due to the secondary laser sputtering just like recombination emissions in the recombination method [6]. These signals can be a guide for searching for unknown transitions of the atoms in liquid helium and can also be a measure of the successful production of isolated atoms in liquid helium.

We have also investigated a dependence of the emission intensity on the probing duration time of the dye laser. The result shows that the Ba atoms escaped in a few tens of milliseconds out of the probe laser beam, the waist of which was about 0.3 mm. The observation time was limited by this and will get longer if we use an unfocused laser beam for probing.

Here we shall give the laser-spectroscopic results so far obtained for the triplet states of Ba and Ca atoms and for the singlet state of Ba atoms in liquid helium (see Fig. 1 for the experimental setup) [15]. We note that the observed spectra did not change when we decreased the power of the sputtering laser and delayed the time for photon counting after sputtering (several milliwatts and several milliseconds) in order to decrease the heating effect. This leads us to believe that the observed spectra originated from the atoms in superfluid helium. We also note that the metastable $5d6p\,^3D_J$ state of Ba and $4s4p\,^3P_J$ state of Ca were initially created by laser sputtering, which indicates the stability of these states even in the liquid helium. Figure 3 shows the excitation and emission spectra of Ba atoms associated with the transition from $5d6s\,^3D_J$ to $5d6p\,^3P_{J'}$. The excitation spectrum is defined for each emission line as the emission intensity as a function of the laser wavelength for excitation, so that it is equivalent to the absorption spectrum in the present case. On the other hand, the emission spectrum is obtained for a particular excitation laser wavelength by scanning the monochromator and plotting the intensity of the emission light with the excitation wavelength fixed. It is noted that the frequencies of the absorption and emission are different in general in liquid helium because the absorption occurs under the helium configuration stabilized for the atomic ground state, whereas the emission occurs after the surrounding helium relaxes to a new equilibrium configuration determined by the excited state wave function. The obtained excitation spectrum [Fig. 3(a)] was very broad (about 7 nm FWHM), asymmetrical, and blueshifted (about 6.5 nm) from the spectrum of the free atom. Figures 3(b) and 3(c) show the corresponding emission spectra for the excitation at (b) 584 nm and (c) 574 nm, respectively. Each emission line is symmetrical and is much narrower (about 1 nm) than the excitation lines. It is slightly blueshifted by about 1 nm. We have thus obtained the excitation and emission spectra for all the transitions between $5d6s\,^3D_J$ and $5d6p\,^3P_{J'}$ states [16]. Some of them were already reported by Bauer *et al.* [7] and our results are in essential agreement with theirs.

We have for the first time observed the spectra for Ca atoms in the triplet state in liquid helium. Figures 4(a) and 4(b) show, respectively, the observed excitation and emission spectra corresponding to the transition between $4s4p\,^3P_J$ and $4s4s\,^3S_1$ states. The excitation spectrum is also very broad (about 7 nm FWHM) and blueshifted (about 6.5 nm) for the spectrum of the free atom. Each emission line is symmetrical and is much narrower (about 0.8 nm) than the excitation line as well.

The information on the optical spectra of atomic Ba and Ca in superfluid helium is quite useful for understanding the structural and dynamical nature of the atomic impurity. Since the observed spectra can be explained rather quantitatively using configuration coordinate diagrams based on an atomic bubble model [7] which was proposed as the structure of the impurity, we can conclude that the model is also valid in these cases.

Furthermore, we can obtain insight for the dynamics of the interaction with surrounding helium. The existence of the emission from all of the $5d6p\ ^3P_{J'}$ states of Ba indicates that the relaxation between the $5d6p\ ^3P_{J'}$ states is quite slow compared with the radiative decay [12].

The advantages of our sputtering method as compared with the recombination method are as follows: (1) Many kinds of atoms can be directly created within liquid helium; (2) only a sample and a pulsed laser are required; (3) it is applicable to pressurized liquid or solid helium; (4) fluorescence signals at the moment of sputtering can be a guide in searching for unknown transitions in liquid helium; and (5) atoms under electric fields can be studied because we need not use them for implanting the atoms. The disadvantages are as follows: (1) The method does not work in the normal state of liquid helium and (2) in spite of the high density, the atoms were created only along the secondary laser beam, so the observation region for spectroscopy was relatively restricted.

Finally, in the process of laser sputtering in superfluid helium we have found a peculiar material produced in the liquid. It was a tangle of thin wires resembling a cobweb or tree. It is surprising that thin wires grew in superfluid helium as a result of laser sputtering. The wire typically had a diameter of less than 1 mm and was electrically conductive, so it might be made of the material of the sample used (Ba or Ca). This phenomenon is not fully understood so far. By careful observation it appeared that the wires grew along the sputtering laser beam while the clusters were laser trapped along the beam. A detailed study of the formation is planned.

This work was partially supported by the Matuo Foundation and the Grant-in-Aid for Scientific Research (02452049,04554010).

[1] See, for example, A. L. Fetter, in *The Physics of Liquid and Solid Helium,* edited by K. H. Bennemann and J. B. Ketterson (Wiley, New York, 1976).

[2] An electron in liquid and solid helium is known to be self-trapped in a large spherical well (about 16 Å radius) due to the large zero-point motion. Recently this "bubble" structure has been unambiguously confirmed in a direct way by observing an optical excitation spectrum of the electron bubble: C. C. Grimes and C. Adams, Phys. Rev. B **41**, 6366 (1990).

[3] R. J. Donnelly, *Quantized Volticies in HeII* (Cambridge Univ. Press, New York, 1991).

[4] M. Iwasaki *et al.*, Phys. Rev. Lett. **67**, 1246 (1991); T. Yamazaki *et al.*, Phys. Rev. Lett. **63**, 1590 (1989).

[5] An exception was the successful implantation of nitrogen atoms and molecules: E. B. Gordon, A. A. Pel'menev, O. F. Pugachev, and V. V. Khmelenko, Fiz. Nizk. Temp. **8**, 601 (1982) [Sov. J. Low Temp. Phys. **8**, 299 (1982)]. Optical spectra of helium atoms and molecules were also studied: W. S. Dennis, E. Purbin, Jr., W. A. Fitzsimons, O. Heybey, and G. K. Walters, Phys. Rev. Lett. **23**, 1083 (1969).

[6] H. Bauer, M. Beau, A. Bernhardt, B. Friedl, and H. J. Reyher, Phys. Lett. A **137**, 217 (1989); H. Bauer, M. Beau, B. Friedl, C. Marchand, K. Miltner, and H. J. Reyher, Phys. Lett. A **146**, 134 (1990); H. Bauer, M. Beau, J. Fisher, H. J. Reyher, J. Rosenkranz, and K. Venter, Physica (Amsterdam) **165-166B**, 137 (1990).

[7] H. Bauer, M. Beau, B. Friedl, C. Marchand, K. Miltner, and H. J. Reyher, Phys. Lett. A **146**, 134 (1990); G. zu Putlitz and M. R. Beau, *Dye Lasers 25 Years,* edited by Michael Stuke (Springer-Verlag, Berlin, 1992).

[8] A preliminary result was reported in Proceedings of the XIII International Conference on Atomic Physics (ICAP), Munchen, 1992 (to be published).

[9] The use of metal as a sample is not essential, for we could successfully implant Ca atoms and observe the signals using CaF_2 crystals doped with La^{2+}.

[10] We have so far used second harmonics (about 200 μJ) and the fundamental (less than about 1 mJ) of a Q-switched and mode-locked Nd:YAG laser (Spectra Physics model 3000) or second harmonics (about 200 μJ) of a Q-switched Nd:YLF laser (Spectra Physics model TFR).

[11] For Ba atoms in liquid helium the excitation and emission wavelengths were already reported for some transitions in Ref. [7].

[12] We have measured lifetimes of $5d6p\ ^3P_J$ states by the correlated single photon counting method, and they turned out to be about the same as those in vacuum (10 ns). The results will be published elsewhere.

[13] The liquid temperature was raised by about 0.2 K with about 2 W of the sputtering laser. With 200 mW the effect was negligibly small.

[14] G. E. Spangler and F. L. Hereford, Phys. Rev. Lett. **20**, 1229 (1968); K. Okuda, Y. Inaba, and M. Date, J. Phys. Soc. Jpn. **34**, 1425 (1973).

[15] These signals has a good S/N ratio, but the signal intensities were rather scattered. This is because the sample acquired holes or pits in the surface during the course of the laser sputtering. Care was required in producing sputtering conditions to obtain the accurate shape of the excitation and emission spectra.

[16] The signals for the transition $5d6s\ ^3D_1 \leftrightarrow 5d6p\ ^3P_2$ were also observed. They were so small that they were not found in Figs. 3(b) and 3(c). The excitation spectrum is about the same as those for the emission lines of $5d6s\ ^3D_2 \leftrightarrow 5d6p\ ^3P_2$ and $5d6s\ ^3D_3 \leftrightarrow 5d6p\ ^3P_3$. The emission spectrum also had the features of the narrow bandwidth (about 1 nm) and the small blueshift (about 1 nm).