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Optical spectroscopy of atoms trapped in solid helium

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We report a study of implantation and optical spectroscopy of foreign atoms in a solid-helium matrix. With a sample of more than 10^9 Ba atoms in 1 cm^3, a storage time in excess of 5 hours has been demonstrated. Laser-induced fluorescence studies favor the assumption that the isotropy of the trapping site for the implanted atoms is preserved during the liquid-to-solid phase transition.

The study of the optical properties of atomic defects in superfluid helium (He II) is becoming a rapidly growing branch of laser spectroscopy.[1-3] Implanted foreign atoms may serve, on one hand, to study the interaction of isolated atoms with a high density helium surrounding. On the other hand, the superfluid helium matrix is a trap for atoms with storage times of up to 10 seconds and densities of typically 10^7-10^8 atoms.[4] The isotropic and diamagnetic surrounding of the implanted atoms makes such a sample attractive for the study of magnetic resonances in paramagnetic atoms which are expected to exhibit extremely narrow line widths. In a former publication we proposed to study optical pumping of paramagnetic atoms in superfluid helium and discussed the possibility to use such atoms to search for *T*- and *P*- symmetry forbidden permanent electric dipole moments.[4] Recently Yabuzaki, Kinoshita, and Takahashi have succeeded in optically pumping Rb and Cs atoms in He II and have observed ground-state magnetic resonance in a double resonance experiment.[5] The short residence time of the atoms in that experiment required the use of a large rf power which considerably broadened the magnetic resonance. The loss of atoms from the observation volume due to diffusion is negligible; in practice however convection currents inside the sample cell originating from temperature gradients limit the observation times. Although we have observed trapping times of Ba atoms in He II of more than 10 seconds,[4] much longer storage times might be achievable with atoms embedded in solid helium.

The structure of a point defect formed in liquid He by a foreign atom is similar to that formed by an electron; both are isolated in "bubbles," i.e., regions from which He atoms are expelled by virtue of the Pauli force.[6,7] Recent spectroscopic studies[8,9] of electrons implanted in condensed helium showed that their optical properties undergo no qualitative changes when implanted in liquid or solid helium. This suggests that the isotropic bubble structure is not changed by the liquid-solid phase transition. The question whether this also holds for atomic impurities has not been addressed so far. In this paper we report the optical spectroscopic experiments on Ba atoms trapped in a He crystal. In our experiments a He crystal was grown by pressurizing over a capillary tube, a copper pressure cell with a volume of approximately 20 cm^3 located inside an optical double bath He cryostat described earlier.[4] The lower part of the cell is in contact with superfluid helium cooled to 1.5 K by pumping He off the liquid surface. Four windows allow optical access to the cell. Under our conditions the liquid-solid phase transition occurs at 27 bar. The crystal was grown as follows. First we raised the pressure in the cell to 26 bar and waited until thermal equilibrium was reached. A subsequent rapid increase of the pressure to 45 bar then triggered the crystallization which takes several seconds. After crystallization was complete the capillary pressure read 33 bar. The crystal was perfectly transparent and no defects could be seen upon visual inspection. The impurity atoms were produced by laser ablation with focused fundamental or second harmonic of a pulsed Nd-YAG laser radiation (typically 40 mJ pulse energy, 3 ns pulse duration, 0.1 mm spot size) from a solid Ba target fixed at the bottom of the pressure cell. The heat deposited during the ablation process locally melts the crystal, which reforms however almost instantaneously after the Nd-YAG laser is turned off. After a few laser shots a conically shaped region of grey color can be seen above the Ba target. Inspection with a long-distance microscope reveals macroscopic particles inside this region which corresponds to the molten portion of the crystal where the particles were distributed by connective flow. When a longer sequence of laser shots is fired onto the target, the defect region grows in the upward direction. The size of the impurity domain used for the following experiments was approximately 1-2 cm^3. We have chosen barium for the present investigation because various studies of its implantation and optical properties in superfluid helium have been reported previously.[3,4,7] The presence of atomic species in this region was detected in a laser-induced fluorescence (LIF) experiment on the $6s^2\,{}^1S_0 \rightarrow 6s6p\,{}^1P_0$ resonance line. This experiment is similar to the one performed earlier in superfluid helium. The beam from a Rh110 ring dye laser tuned by a computer controlled birefringent filter traverses the impurity region in the pressure cell and fluorescence light is collected at 90 degrees. It is analyzed by a monochromator and a photon counting system. The laser beam had a diameter of 3 mm and the power at the entrance of the cryostat was 1 to 6 mW. The implantation procedure described above proved to be reproducible, although the detected LIF count rates could vary by up to one order of magnitude in different experimental cycles (crystal growth and implantation). The maximal observed LIF count rate was as high as 600 kHz. With an overall detection efficiency of our apparatus of 10^{-5} and an estimated absorption cross section of 10^{-15} cm^2 this

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corresponds to a number density of implanted atoms of 3×10^9 cm^{-3}.

A typical line shape of the emission spectrum with the excitation wavelength set to 542 nm is shown in Fig. 1(c). Solid lines are drawn to guide the eye. The central wavelength of the Ba fluorescence line is blueshifted by 1.9(5) nm with respect to the free atomic line (553.6 nm) and has a width (FWHM) of 1.3 nm. For comparison we also show in Figs. 1(a) and 1(b) emission spectra recorded in He II at pressures of 1.2 and 25.6 bar. The exciting wavelengths were 548.1 and 546.0 nm, respectively. The asymmetric background and the additional small peaks which can be seen on the spectra obtained in liquid He are due to scattered laser light and Raman scattering in the fused silica windows. When the exciting wavelength is tuned, these Raman lines show an equivalent detuning, while the Ba fluorescence line remains unshifted and shows merely a decreased amplitude. In the case of atoms trapped in solid He, the count rate was two orders of magnitude larger, and no contribution from the scattered light could be seen.

Excitation spectra were recorded by scanning the dye laser wavelength and recording fluorescence at the peak of the emission line. Typical line shapes (normalized to laser power) recorded in solid [2(c)] and liquid [2(a) and 2(b)] helium are shown in Fig. 2. The solid lines are again drawn to guide the eye. The emission line is slightly broadened when going from liquid to solid helium, whereas the excitation line shows no significant change in shape. This behavior may serve as a sensitive test in model calculations of the optical properties of atoms implanted in condensed helium. To our knowledge there is at present no quantitative theory describing these properties. A preliminary investigation of the pressure shift of the Ba excitation line in superfluid He yielded a shift parameter of -0.14 ± 0.03 nm/bar with an extrapolated barycenter at 0 bar of 545.5 nm. If we assume that this linear pressure shift observed in the liquid phase can be extrapolated into the solid phase, the measured line shift in solid helium would correspond to a pressure of 30 bar consistent with the applied pressure.

A systematic study of the dependence of the fluorescence signal on laser power was impeded by the onset (at 30 mW) of strong Schlieren formation around the laser beam emerging from the cryostat, accompanied by a rapid decrease in the fluorescence count rate. Laser light absorption by macroscopic impurities led to a local melting of the He crystal with a subsequent loss of Ba atoms by cluster formation. Although the samples used in the described experiments were stable on the time scale for several hours, some decrease in the fluorescence counting rate could be observed already at lower power (several mW). We attribute the latter to the laser melting process.

We have shown that a large number of atomic defects can be implanted into a solid helium matrix, where they may be trapped for many hours. The excitation and emission spectra of Ba atoms in solid helium reported here indicate that the liquid-solid phase transition, as for electrons, does not cause a qualitative change in the optical properties. This fact leads us to conclude that the isotropic spherical surrounding (bubble structure) seen by Ba atoms in He II is not noticeably changed in the solid matrix, in contrast to heavier noble gas matrices where crystalline fields and the dynamic Jahn-Teller effect lead to a strong broadening and splitting of the resonance lines of implanted atoms.[10,11]

Solid helium has proven to be an efficient trap for

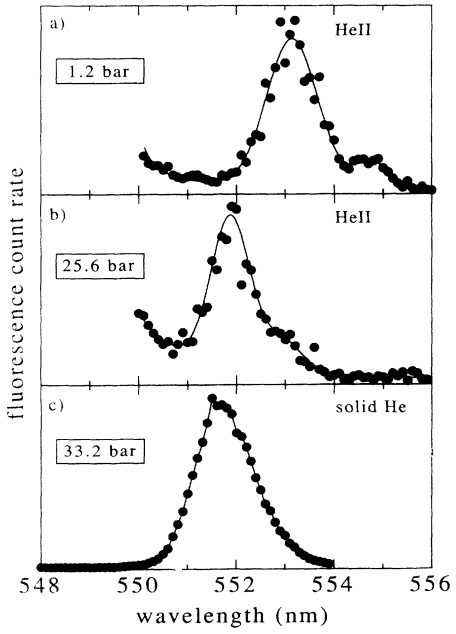

FIG. 1. Emission spectra of Ba atoms implanted in superfluid helium at a pressure of 1.2 bar (a), 25.6 bar (b) and trapped in a solid helium matrix at 33.2 bar (c). The excitation wavelengths were 548, 546, and 542 nm, respectively.

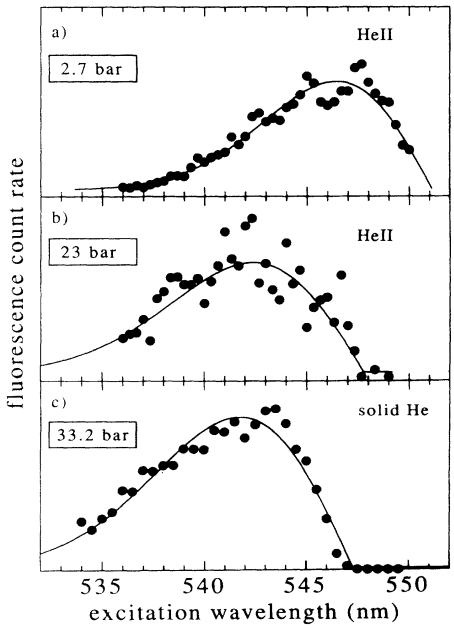

FIG. 2. Excitation spectra of Ba atoms implanted in superfluid helium at a pressure of 1.2 bar (a), 23 bar (b) and trapped in a solid helium matrix (c).

foreign atoms. Although the helium trap is too perturbative to allow high resolution optical spectroscopy it offers the potential capability for an optical detection of narrow ground-state magnetic resonance signals in paramagnetic atoms. As relaxation processes arising from spin-spin interactions in such dilute samples are negligible, the long interaction time of the sample with external fields is expected to yield low longitudinal and transverse relaxation rates. Experiments aimed at measuring the latter are underway in our laboratory.

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[1]M. Himbert, A. Lezama, and J. Dupont-Roc, J. Phys. **46**, 2009 (1985).

[2]G. zu Putlitz and M. R. Beau, in *Dye Lasers: 25 Years,* edited by M. Struke (Springer, Berlin, Heidelberg, 1992).

[3]T. Yabuzaki, A. Fujisaki, K. Sano, T. Kinoshita, and T. Takahashi, *Atomic Physics 13,* edited by H. Walther and T. W. Hänsch (North-Holland, New York, 1992).

[4]M. Arndt, S. I. Kanorsky, A. Weis, and T. W. Hänsch, Phys. Lett. A **174**, 298 (1993).

[5]T. Yabuzaki, T. Kinoshita, and Y. Takahashi, in *Laser Spectroscopy XI,* edited by L. Bloomfield, T. Gallagher, and D. Larson (AIP, New York, 1993); T. Kinoshita, Y. Takahashi, and T. Yabuzaki, following paper, Phys. Rev. B **49**, 3648 (1994).

[6]A. Lezama, these, Université P. et M. Curie (Paris VI), 1981.

[7]H. Bauer, M. Beau, B. Friedl, C. Marchand, K. Miltner, and H. J. Reyher, Phys. Lett. A **146**, 134 (1990).

[8]C. C. Grimes and G. Adams, Phys. Rev. B **41**, 6366 (1990).

[9]A. I. Golov and L. P. Mezhov-Deglin, Pis'ma Zh. Eksp. Teor. Fiz. **56**, 527 (1992) [JETP Lett. **56**, 514 (1992)].

[10]S. L. Kupferman and F. M. Pipkin, Phys. Rev. **166**, 207 (1968).

[11]H. Kuppelmaier, H. J. Stöckmann, A. Steinmetz, E. Görlach, and H. Ackermann, Phys. Lett. **98A**, 187 (1983).