Measurement of the infrared absorption spectrum by negative charges in solid helium

A. I. Golov and L. P. Mezhov-Deglin
Institute of Solid State Physics, Russian Academy of Sciences, 142432 Chernogolovka, Moscow Province, Russia

(Submitted 22 October 1992)
Pis'ma Zh. Eksp. Teor. Fiz. **56**, No. 10, 527–531 (25 November 1992)

Absorption of infrared radiation by negative charges in solid helium has been observed. The pressure dependence of the line position, and also its width and absorption cross section, have been measured. Experimental confirmation of the existing bubble model of the electron in solid helium has been obtained. The values of various parameters of this model are determined and discussed.

The properties of charges in solid helium have been studied now for more than thirty years.[1] Starting with the first papers of Shal'nikov,[2] researchers have been interested mainly in the transport properties of these charges. The structure of negative charges in solid helium has not been experimentally investigated. By analogy with liquid helium, it has been assumed[1,3,4] that the electron in solid helium is localized in a spherical cavity of radius on the order of 10 Å. Atoms of helium are expelled from this cavity as a consequence of the short-range repulsion.[5,6] However, until now this model has not been experimentally verified, and those properties which may be due to the influence of the structure of the crystal matrix on the shape of the cavity have not been studied.[7] Studies of the properties of the analogous defect-positronium in solid helium, have shown[8] that the effective radius of the bubble around the positronium varies from 10 to 8 Å as the solidification pressure is increased from 40 to 100 atm.

The bubble model, which has made it possible to explain the long lifetime of positronium in liquid helium, was proposed by Ferrell 35 years ago.[3] Since then, its ability to describe the properties of the electron in liquid helium has been confirmed by various methods.[4] It has turned out that the very simple model of a spherically symmetric, rectangular potential well is adequate. The main parameter in such a problem is the radius of the well (of the bubble), which varies from 17 to 12 Å as the pressure is varied between 0 and 25 atm. In such a potential well the electron has discrete levels. This permits one to formulate the problem experimentally as an investigation of the properties of negative charges in helium by spectroscopic methods. Calculations of the optical properties of electrons in liquid helium are presented in Refs. 9 and 10. Recently, the first measurements were made of absorption of infrared radiation during the transition of the electron from the $1s$ ground state to the $1p$ excited state in liquid helium.[11,12] These experiments have confirmed the model.[3]

Detection of the $1s$–$1p$ absorption line of negative charges in solid helium would unambiguously confirm the applicability of the bubble model. The goal of the present effort is to observe absorption of light by negative charges in solid helium.

The working cell—a flat diode (electrode dimensions 6×20 mm, gap between the radioactive sources and the collector $L=0.8$ mm)—was placed inside a metal cell with sapphire windows. Crystals of solid helium were grown in the cell from compressed liquid at constant pressure from above downward from a copper heat sink to the capillary of the filling system. Growth of the crystal was monitored by the drop in the collector current I at constant voltage U between the charge source and the collector. The temperature of the cell was measured by a carbon rheostat thermometer, mounted on the outer surface of the cell. A temperature gradient was maintained along the cell during the growth of the crystal. Because of the large thermal conductivity of the metal walls, the cell sometimes became blocked during the final stage of growth of the sample. For this reason, the values of the solidification pressures given below may be slightly higher.

The degree of homogeneity of the samples was estimated from the transient characteristics of the collector current upon incremental application of a constant voltage. For the optical investigations we chose samples with a sharply defined leading edge in the transient characteristic.

Radiation from a quartz halogen lamp was bent with the help of a prism monochromator (spectral width ~ 0.07 μm) into an interval of 3–5 μm and focused in the helium crystal between the faces of the diode. Immediately after exiting the cell the light falls upon a carbon bolometer cooled to a temperature of 1.5 K. The lamp light was modulated by a shutter with a frequency of 185 Hz. The modulated signal from the bolometer, proportional to the total power of the light passing through the diode, was amplified by an external selective amplifier with synchronous detector and then recorded.

In the first experiments the concentrations of the negative charges in the gap between the faces of the diode were modulated by alternatively turning the negative voltage on and off. For small mobilities of the charges, when the current is small in comparison with the saturation current of the β-source (20 nA in our diode), the collector current is limited by the space charge. In the space-charge-limited current (SCLC) region the mean charge density n is proportional to the applied voltage. The charge mobility in solid helium on the melting curve is roughly three orders of magnitude smaller than in the liquid and falls off exponentially from there as the temperature is lowered. The SCLC region in solid helium therefore extends to fields of the order of 10^4 V/cm. This allowed us to make spectroscopic measurements in solid helium at voltages of a few kilovolts, i.e., for charge densities 2–3 orders of magnitude higher than in the liquid in our experiments.[11,12]

We found that the maximum (steady-state) absorption signal was reached after 2–3 transit times by the negative charge front of the source-collector interval. Light absorption by the charges is proportional to the difference in the bolometer readings with the source voltage turned on and off. In the first experiments this quantity was determined by digital subtraction of the voltage, $U=-4.5$ kV. To improve the signal-to-noise ratio, the measurement process at fixed wavelength was repeated from 100 to 800 times, and the difference in the readings was summed. The recording of one spectrum ordinarily took from three to ten hours. Control of the experiments and recording and processing of the data were automated with the help of a personal

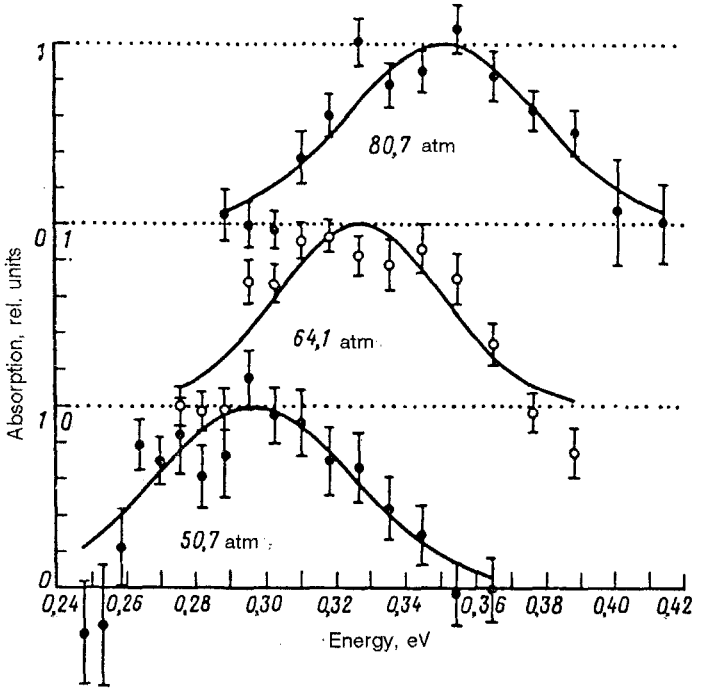

FIG. 1. Diagram of the absorption of negative charges in solid ^{4}He for three different pressures. For convenience, the absorption curves have been staggered along the vertical axis. The vertical line segments indicate the limits of measurement error.

computer and CAMAC interface.

As our experimental results have shown, turning on a positive voltage of roughly the same magnitude makes it possible to greatly speed up the departure of negative charges from the volume after turning off the negative voltage. This procedure allowed us to significantly shorten the time required to complete all the measurements. The optimal parameters—those for which it was possible to achieve a relative value of the absorption of light by the electrons greater than 0.001—were as follows: negative voltage $U=-4.5$ kV (electric field 5×10^4 V/cm), positive voltage $U=+4$ kV, collector current 1.4 nA, transit time of the working interval by the negative charge front 1.5—2 sec, charge mobility 10^{-6} cm^2/(V · sec), time interval between switching on of the electric field 5—8 sec. Here the negative charge density in the volume was close to 6×10^{11} cm^{-3}. Since the charge mobility on the melting curve in hcp ^{4}He is of the order of 10^{-5} cm^2/(V · sec), to satisfy the above-enumerated conditions it was necessary to work at temperatures 0.3–0.7 K lower than the melting point. The described procedure for determining the absorption is, in fact, a digital synchronous detection at frequencies of the order of 0.1 Hz.

Figure 1 shows the IR absorption spectra in three crystals of hcp ^{4}He, grown at

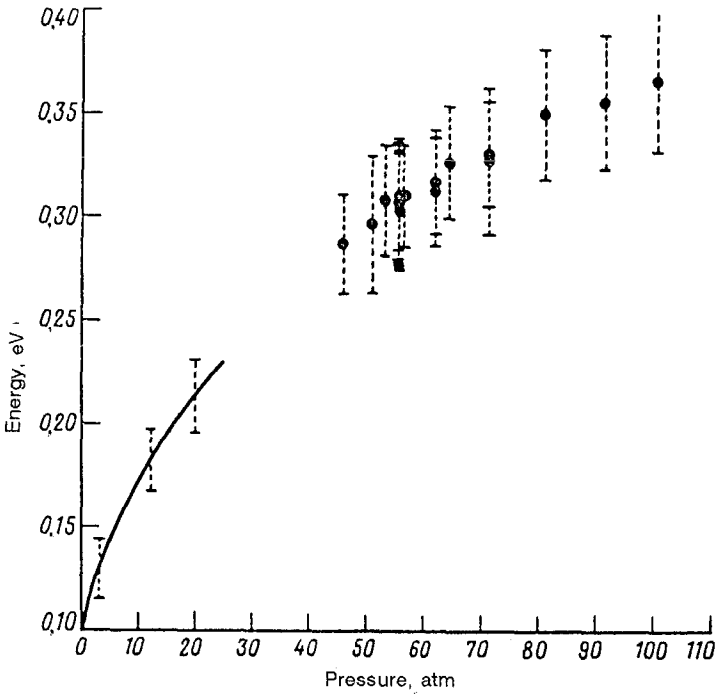

FIG. 2. Dependence of the energy of the $1s$–$1p$ transition on the pressure in solid ^{4}He. The dashed lines indicate the width of the absorption line at half-height. The solid curve was constructed from results of analogous measurements[11] in liquid helium.

different pressures. The vertical line segments indicate the limits of statistical error. Each spectrum was approximated by a Gaussian function, from which we extracted the position of the center of the E_{1s-1p} line (and from that the energy of the $1s$–$1p$ transition) and its width. Within the limits of accuracy of our experiments and the accessible range of temperatures, we were not able to observe any dependence of the position or width of the line on the temperature or the applied voltage.

Figure 2 shows the dependence of the energy of the $1s$–$1p$ transition on the pressure in both liquid and solid ^{4}He. The dashed vertical line segments indicate the line widths at half-height. The solid line was constructed from measurement data[11] in liquid helium. The points are our measurements in solid helium.

To determine the bubble radius from the magnitudes of E_{1s-1p} in the rectangular spherical potential well model, it is sufficient to know its depth (see, e.g., Ref. 13). As in Ref. 5, we calculated it by the Wigner–Seitz method,[14] which gave good agreement with the results of an experimental measurement study in which an electron was introduced into liquid helium and dense gaseous helium.[15] On the basis of our estimates, in the pressure range 46–100 atm the bubble radius decreases continuously from 10.7 to 9.4 Å.

Unfortunately, we do not know of any theoretical calculations of the line width and absorption cross section of the $1s$–$1p$ transition in solid helium. For the case of liquid helium such calculations were performed in Refs. 9 and 10. Measurements of the absorption in the liquid have shown[11,12] that the line width is roughly two times greater than the estimates,[9,10] which is roughly six times smaller than the transition energy E_{1s-1p}. More recent calculations of the properties of an electron in liquid helium by the Monte Carlo method[16] have confirmed the applicability of the simple model[3] for finding the bubble radius, but give a substantially wider absorption line for the $1s$–$1p$ transition. In our measurements the line width in solid helium is also 5–6 times smaller than the transition energy.

The digital method of determining the absolute value of the total absorption allowed us to estimate the absorption cross section. The negative charge density in the volume was determined independently from known values of the collector current and the velocity (the transit times). This cross section was 12 ± 3 Å^2 over the entire pressure region. Note that the found value of the cross section is near the calculated value of 21 Å^2 for an electron in liquid helium at pressures of 0–20 atm,[10] obtained by assuming that the line width is equal to 0.1 eV.

We thank A. Lokhov, E. N. Solov'ev, and Ya. E. Volokitin for their help in preparing the equipment and carrying out the experiments, and also to S. V. Pereverzev for useful discussions.

[1] A. J. Dahn, in: Progress in Low Temp. Phys., ed. by D. F. Brewer, North-Holland, Amsterdam, Vol. 10, 1986, p. 73.
[2] A. I. Shal'nikov, Zh. Eksp. Teor. Fiz. **47**, 1727 (1964) [Sov. Phys. JETP **20**, 1161 (1965)].
[3] R. A. Ferrell, Phys. Rev. **108**, 167 (1957).
[4] K. W. Schwarz, in: Advances in Chem. Phys. XXXIII, ed. by L. Prigogine and S. A. Rice, Wiley, New York, 1975, p. 1.
[5] M. H. Cohen and J. Jortner, Phys. Rev. **180**, 238 (1969).
[6] V. B. Shikin, Zh. Eksp. Teor. Fiz. **61**, 2053 (1971) [Sov. Phys. JETP **34**, 1095 (1972)].
[7] A. I. Golov, Pis'ma Zh. Eksp. Teor. Fiz. **49**, 346 (1989) [Sov. Phys. JETP Lett. **49**, 396 (1989)].
[8] J. B. Smith, J. D. McGervey, and A. J. Dahm, Phys. Rev. B **15**, 1378 (1977).
[9] I. A. Fomin, Pis'ma Zh. Eksp. Teor. Fiz. **6**, 715 (1967) [Sov. Phys. JETP **6**, 196 (1967)].
[10] T. Miyakawa and D. L. Dexter, Phys. Rev. A **1**, 513 (1970).
[11] G. C. Grimes and G. Adams, Phys. Rev. B **45**, 2305 (1992).
[12] A. Ya. Parshin and S. V. Pereverzev, Zh. Eksp. Teor. Fiz. **101**, 126 (1992) [Sov. Phys. JETP **74**, 68 (1992)].
[13] L. F. Schiff, *Quantum Mechanics*, McGraw–Hill, New York (1955).
[14] J. Jortner, N. R. Kestner, S. A. Rice, and M. H. Cohen, J. Chem. Phys. **43**, 2614 (1965).
[15] J. R. Broomall, W. D. Johnson, and D. G. Onn, Phys. Rev. B **14**, 2819 (1976); U. Asaf and I. T. Steinberg, Chem. Phys. Lett. **128**, 91 (1986).
[16] D. F. Coker and B. J. Berne, J. Chem. Phys. **89**, 2128 (1988).

Translated by P. F. Schippnick