Relative momenta are less than k_F which is 0.3 Å⁻¹ for a 6% mixture and so the D-state impact parameter lies outside the range of the interparticle force. Thus the scattering amplitude is dominated by S and P states and we may take $A_E(\mathbf{k}, \phi)$ to be $A_E(\mathbf{k}, 0)$ and $A_0(\mathbf{k}, \phi)$ to be $A_0(\mathbf{k}, 0)\cos\phi$. Measurement of χ and D as functions of concentration and hence k_F at temperatures of the order of 0.01°K would give $A_E(\mathbf{k}, 0)$ and $A_0(\mathbf{k}, 0)$, provided rearrangement effects²² could be estimated. The method of obtaining a fermion superfluid transition temperature T_c from the scattering amplitudes is given in Ref. 18.

Present indications^{23,24} are that χ is very close to χ_0 for concentrations of a few percent so that the right-hand side of Eq. (4.1) is small. This result does not necessarily imply that the effective forces are weak since $A_0(\mathbf{k},0)$ and $A_E(\mathbf{k},0)$ can cancel each other on average. In addition, the scattering amplitudes reflect the behavior of the potential and change sign as k

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24 A. C. Anderson and D. O. Edwards (private communication).

increases so that the integrals of $A_0(\mathbf{k},0)$ and $A_E(\mathbf{k},0)$ can separately be small even though the amplitudes themselves are large for some values of k.

However, measurements of the spin diffusion in dilute mixtures²⁴ show that the lifetimes of the He³ quasiparticles are 50–100 times larger than in pure He³. It is not easy to attribute this to cancellations in the right-hand side of Eq. (4.3) and so the scattering amplitudes appear to be much smaller in magnitude than in pure He³. Detailed estimates, based upon the value of D only, depend upon assumptions about the relative values of the S- and P-state scattering amplitudes. Nevertheless it seems unlikely that dilute mixtures will undergo a fermion superfluid phase transition at presently attainable temperatures.

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Behavior of Ions and Quantized Vortex Rings of Low Energy in Helium II around 0.3°K

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Quantized vortex rings of low energy down to 0.1 eV have been produced in specially purified liquid helium at 0.3° K and the deflection of rings of 0.15- to 0.5-eV energy in a transverse magnetic field measured. In the same apparatus the Hall mobility of positive ions has been measured at 0.3° K, yielding after extrapolation to zero electric field a value $\mu = 3.0 \times 10^4$ cm²V⁻¹ sec⁻¹. The drift velocity of ions can reach 40 m sec⁻¹ before vortex rings are produced, also at 0.3° K.

INTRODUCTION

 ${f R}$ ECENTLY, Rayfield and Reif¹ have shown that ions in He II, accelerated beyond thermal velocity, produce vortex rings of one quantum of circulation. They studied the behavior of these rings in detail in the energy range of 5–50 eV, the rings having a radius between about 0.2–2 μ . Even if the circulation of these rings is quantized, their hydrodynamic behavior is essentially classical.² Therefore, it seemed of interest to make rings of as low an energy as possible with correspondingly small radius, in the hope of getting some insight into the behavior of the rings as they approach the quantum hydrodynamic region.

This should also yield information on the mechanism producing such rings.

The energy E of a vortex ring of radius R (assuming the core with radius a is solid) is given by²⁻⁶

$$E = \frac{1}{2}\rho\kappa^2 R \left[\eta - 7/4 \right], \tag{1}$$

where ρ is the density of the fluid; κ , the circulation, is h/m (*m* is the mass of the He atom) in this case; and $\eta = \ln 8R/a$.

Since each part of the vortex ring is in the velocity field of the other parts, a net velocity v results, perpen-

²² N. M. Hugenholtz and L. Van Hove, Physica 24, 363 (1958). ²³ D. L. Husa, D. O. Edwards, and J. R. Gains, Bull. Am. Phys.

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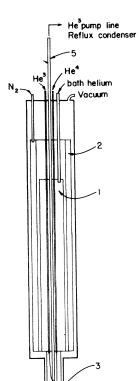


Fig. 1. Schematic drawing of cryostat.

dicular to the plane of the ring

$$v = (\kappa/4\pi R)(\eta - \frac{1}{4}). \tag{2}$$

Table I contains the energy and velocity of vortex rings with one quantum of circulation, according to Eqs. (1) and (2), for rings of 10-200-Å radius. We assume a core of radius 1 Å. Table I shows clearly that vortex rings with one quantum of circulation have a radius approaching atomic dimensions only if the energy of the rings is 0.1 eV or less. Then the quantity in the brackets in Eq. (1) or (2), depending on the model used for the core, is no longer only a slowly varying function of R and cannot be treated as a constant.

We were able to make vortex rings of 0.1 eV, corresponding to a radius of about 50 Å and a velocity of 900 cm/sec, and also to study the deflection of vortex rings of 0.15–0.5 eV by a magnetic field. The same equipment was used to measure the Hall mobility of positive ions at $0.3\,^{\circ}\mathrm{K}$.

METHOD AND APPARATUS

The apparatus was described briefly in a preliminary note.⁷ It is shown schematically in Fig. 1. A cell (6),

Table I. Energy E and velocity v of a classical vortex ring of radius R and 1-Å radius of a solid core. E^* is the energy of a vortex ring of radius R with the bracket of Eq. (1) replaced by $[\ln 8R/(0.47 \text{ Å})-1.67]$ according to Ref. 2.

<i>R</i> (Å)	<i>E</i> (eV)	v (m/sec)	E* (eV)
20	0.030	19	0.037
40	0.073	11	0.088
60	0.121	7.8	0.142
80	0.171	6.2	0.200
100	0.224	5.1	0.265
120	0.278	4.6	0.324
140	0.331	4.2	0.386
160	0.382	3.8	0.452
180	0.434	3.3	0.518
200	0.512	2.8	0.586

containing about 100 cm³ of liquid He, was mounted between the 4-in. pole pieces (4) of a Varian magnet that produces a maximum reversible magnetic field of 15 000 oersted. The ions were produced by the α particles emitted by a Po²¹⁰ source plated on a holder with a rectangular surface. A grid at a distance of 2-6 mm from the source accelerated the ions produced by the α particles. The grid also had a rectangular form of the same size as the source, in order to produce a collimated rectangular ion beam of 2×10 mm, with its longer side parallel to the magnetic field. The collector had the shape shown in Fig. 2(a); the central part was grounded and the two sides connected to Cary vibrating-reed electrometers. The grounded center part was identical in size to the source surface and the collimating grid. The total grid arrangement is shown schematically in Fig. 2(b). It contained an auxiliary grid 1 mm in front of the collector which was used to distinguish between ions and vortex rings, and to measure the energy loss of the rings in a manner similar to that described by Rayfield and Reif.1 The main drift space between collimator and collector was 1.32 cm long in most experiments, and was free of electric fields for the study of vortex rings.

The cell was cooled by a He³ refrigerator as described by Reif and Meyer.8 A novel feature of this cryostat was the use of the center tube (5) (Fig. 1) for a dual purpose: After the cell was filled with liquid He and cooled down below the λ point, it acted as a pumping tube for the He³ refrigerator. However, during precooling and condensation of the He it was used as a reflux condenser with He4 as exchange gas. When starting an experiment, the nitrogen shield (2) and the main He space were both filled with liquid N₂ and the center tube filled with 1 atm of He4 gas of high purity. Because of thermal convection, the carbon thermometer in the cell (6) read N₂ temperature after about 4 h. We usually precooled to N₂ temperature during the night before a run. The liquid N2 was then removed from space 1, the space flushed with He gas

⁷ L. Meyer, in *Proceedings of the Ninth International Conference on Low Temperature Physics*, edited by J. G. Daunt, D. O. Edwards, F. J. Milford, and M. Yaqub (Plenum Press, Inc., New York, 1965), p. 338.

⁸ F. Reif and L. Meyer, Phys. Rev. 119, 1164 (1960).

and filled with liquid He. Helium gas was added to tube 5 in order to compensate for the increase in density due to the cooling, and to keep the pressure at 1 atm. Pumping on the main He bath (1) reduced its temperature to about 2.2°K and condensed about 1 cm³ of liquid out of the exchange gas in the center tube (5). This liquid ran down towards the evaporator (3) of the He³ refrigerator, evaporated there, and the vapor rose again and recondensed in heat exchange with the main bath (1). This reflux condenser action cooled the cell between the magnet poles from N₂ temperature to the temperature of the bath in about 20 min. Helium gas, purified by a charcoal trap at liquid-N2 temperature, could then be condensed into the cell. This required another 15 min. The temperature of the main bath was then reduced to 1.8-2.0°K and the cell followed almost instantaneously. The He⁴ was pumped out of tube 5 to a pressure less than 10⁻⁵ mm Hg which was usually achieved in less than $\frac{1}{2}$ h. The bath temperature was then lowered to 1.2°K and the He³ system started bringing the cell to about 0.3°K.

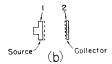
The cell was protected by two coaxial radiation shields of $\frac{1}{16}$ -in. copper covered by at least ten layers of "super-insulation" (Al-covered 0.0005-in.-thick Mylar). One shield was in thermal contact with the He bath and the other (not shown in Fig. 1) with the bottom of the N_2 space. The He bath and N_2 jacket were also wrapped in super-insulation.

The temperature was measured with a Speer carbon resistor of about 150 Ω at room temperature and about 750 Ω at 0.3°K. The thermometer was calibrated against the He³ vapor pressure (cf. Ref. 8), with a slight extrapolation to 0.3°K. Voltages were measured with a Hewlett-Packard vacuum tube voltmeter calibrated with a Wenner potentiometer against a standard cell.

The experiments were performed in the following way: After the cell was filled with liquid He and cooled to the lowest temperature we could reach (0.29–0.30°K), the potential of the source was raised from 0.1 to 0.6 V, without any potential drop across the main drift space. We then measured the current received by the two parts of the collector as a function of the opposing potential on the auxiliary grid in front of the collector. Our observations were similar to those



FIG. 2. Schematic drawing of electric system. (a) Collector surface, (b) grid arrangement.



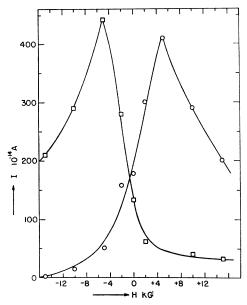


Fig. 3. Currents measured by the two parts of the collector as a function of the magnetic field. The potential between source and collimator is +0.3 V; between auxiliary grid and collector it is -0.02 V.

of Rayfield and Reif.1 We needed an opposing potential of the same amount as the accelerating potential at the source in order to cut off the current. However, the cutoff for vortex rings of very low energy was much less sharp than that reported in Ref. 1, in spite of the fact that the energy loss due to collisions with rotons, photons, and He3 should be negligible at these temperatures. (Cf. Ref. 1, p. 1204.) These measurements were repeated as a function of the magnetic field H; sometimes, however, we used only a fixed opposing potential of 0.02 V, just to eliminate the possibility that an ion current was present besides the current carried by vortex rings. We evaluated only runs where the current-versus-magnetic-field curves were symmetrical for the two collectors, as in the example shown in Fig. 3. The geometry of the cell was such that moving the maximum intensity of the current from one collector to the other produced a deflection of the beam by 0.41 rad.

RESULTS

We encountered unexpected difficulties in making vortex rings of less than 0.5-eV energy. In the first few runs after the system had been open to the air, we found that a relatively higher minimum potential between source and first grid was needed to produce a vortex ring current that could be stopped only by an opposing potential equal to the accelerating potential. This fact suggested that impurities might play a role in this difficulty (cf. Ref. 9). Therefore, we

⁹ J. Jortner, L. Meyer, S. A. Rice, and E. G. Wilson, Phys. Rev. Letters 12, 415 (1964).

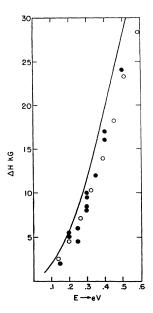


Fig. 4. The magnetic field producing a deflection of vortex rings of 0.41 rad as a function of vortex ring energy. Solid line: Eq. (1) with core of radius 1 Å. Solid circles: Experimental results. Open circles: Eq. (4.14) of Ref. 2.

thoroughly flamed the charcoal trap that cleaned the experimental He after every few runs, and resorted to a purification technique that had proved very efficient for liquid argon.¹⁰ We electrolyzed the liquid He at $0.4\text{--}0.5\,^{\circ}\text{K}$ for several hours with a current of 10^{-11} A produced by a field of 50 V/cm, and reversed the polarity several times. After that treatment we were able to observe positively charged vortex rings with a potential difference as low as 0.1 V between source and first grid, corresponding to a radius of about 55 Å. Measuring the collector current as a function of the opposing potential, we found several times that we needed almost 0.11 V to cut off the current completely. Unfortunately, we did not succeed in measuring the magnetic deflection of the vortex rings below 0.15-eV energy because changing the magnetic field caused eddy-current heating in the cell walls and temporary temperature fluctuations which apparently could sweep impurities back into the main drift space by heat flush, thus destroying the current of low-energy vortex rings. Re-electrolyzing made the vortex current reappear, but such a method was too cumbersome to vield reliable data.

Unfortunately, the error in the experiment is not determined by the accuracy with which we could measure potentials, ion currents, magnetic fields, etc., but rather by the back-diffusion of impurities into the drift space, and by temperature fluctuations during changes of the magnetic field, which are difficult to estimate quantitatively. We discarded all data where the current received by the two collectors was not approximately equal in zero field, and where the current measured by the vibrating-reed electrometers changed with time when the potentials, temperature, and magnetic field were kept constant.

The results are summarized in Fig. 4. The changes in magnetic field, ΔH , necessary to move the current peak from one collector to the other (0.41 rad) is plotted against the accelerating potential used to produce the vortex rings.

We also measured the Hall mobility of positive ions at 0.3° K in a uniform field of 0.166 V/cm, as described in Ref. 7, which yielded an apparent mobility $(2\pm0.2)\times10^{4} \text{ cm}^{2}/\text{V}$ sec, and a drift velocity of $\sim35\times10^{3} \text{ cm/sec}$. Using the universal curve of Ref. 8, the zero-field mobility is $3.0\times10^{4} \text{ cm}^{2} \text{ V}^{-1} \text{ sec}^{-1}$.

We were unable to produce negatively charged vortex rings of low energy in our equipment. This is not in contradiction to the results of Rayfield and Reif¹ who found that vortex rings are created by positive and negative ions. The reason is simply that the drift velocity of the negative ions at these low temperatures is almost two orders of magnitude smaller than that of positive ions (cf. Ref. 8). A potential of 0.1 V between source and first grid ($\sim 0.5 \text{ V/cm}$) is enough to accelerate positive ions with a mobility of $3.0 \times 10^4 \text{ cm}^2 \text{ V}^{-1} \text{ sec}^{-1}$ to more than the 40 m/sec necessary to produce vortex rings. In the same field, negative ions only reach about 2 m/sec and are unable to create vortex rings.

DISCUSSION

The deflection of a beam of vortex rings in a transverse magnetic field can be derived from an impulse approximation, following Ref. 1. If θ is the angle of deflection, it is given by

$$\theta = \Delta p_x / p_0 \,, \tag{3}$$

where Δp_x is the transverse momentum acquired from the magnetic field during the transit of the vortex ring throughout the drift space, and p_0 is the impulse the ring acquired in the electric field from the acceleration of the charge it carries. The transverse momentum Δp_x is the product of the Lorentz force, $(e/c)(\mathbf{v}_0 \times \mathbf{H})$, and the time spent in the drift space L_z/v_0 , where L_z is the length of the drift space and v_0 is the velocity of the ring in the direction of the electric field. This product yields

$$\Delta p_x = (e/c)L_z H. \tag{4}$$

Note that the velocity of the ring v_0 cancels. The deflection angle is then

$$\theta = (e/c)L_z H/p_0. \tag{3a}$$

Using

$$p_0 = \pi \rho \kappa R^2 \tag{5}$$

for the impulse of a vortex ring (see Refs. 6 and 11), we can calculate the field ΔH necessary to deflect the beam of vortex rings with radius R by 0.41 rad through a total length of drift space of 1.67 cm. In order to compare the calculated values of ΔH with the experi-

¹⁰ H. Schnyders, S. A. Rice, and L. Meyer, Phys. Rev. Letters 15, 187 (1965).

¹¹ C. C. Lin, *Liquid Helium*, edited by G. Careri (Academic Press Inc., New York, 1963).

mental data, we have to know the energy E as a function of radius R. Rayfield and Reif¹ use Eq. (1) as it stands and derive a value for the core radius of a =1.28 Å from their data. Using the same values, we get the drawn line in Fig. 3, which deviates systematically from the experimental data. The calculated fields are systematically too high outside our experimental error. Parks and Donnelly12 prefer 1.46±0.14 Å for the core radius, subtracting 1.6216 instead of 7/4 in Eq. (1), which would slightly increase the discrepancy. All corrections to the geometry of the apparatus (for end effects, etc.) would also only increase the discrepancy, since θ would increase, and, therefore, the calculated ΔH , since it is proportional to θ . Recently, Amit and Gross² gave a variational treatment to the quantum-mechanical approach to the problem of the core of a vortex in superfluid He,3-5 where the core size is essentially given by the healing length of the He wave function. Their value for the bracket in Eq. (1) becomes $[\ln 8R/a - 1.67]$, with a reduced to 0.47 Å. Using Amit and Gross' values in the bracket, we obtain the points, shown as the open circles, that fall in the middle of the experimental data.

It is difficult at the moment to decide whether or not the difference between our results and the values calculated using Eq. (1) in its standard form is really decisive, and whether this can be considered a proof of the correctness of the values derived by Amit and Gross.

The potentials we used were rather small, and surface layers on the electrodes could easily shield part of the applied voltage (to the order of 0.05 V), which would bring our experimental data into agreement with the standard values for the core radius. However, our electrodes were gold-plated and clean, and we never observed the type of erratic results that dirty surfaces produce. We changed the geometry of the drift space and the distance between source and first collimating grid by a factor of 3, without getting any different results. At 0.3°K, the energy loss of the vortex rings in well helium due to scattering by phonons, rotons, and He3 is negligible, as shown by Rayfield and Reif,1 and consistent with our own observations.

The deflections of more energetic vortex rings in an electric field were measured by Rayfield and Reif (Ref. 1, footnote 27) and show a deviation of the calculated values from the experimental data that seem to be of the same sign and the same order of magnitude as the one we observe. However, the discrepancies in both cases are just barely outside the experimental error.

We intend to rebuild the apparatus for use with a He³-He⁴ dilution refrigerator^{13,14} in order to reach 0.1°K by continuous cooling. We also intend to improve our purification to remove the He³ from the He used in the experimental cell. We expect to carry the investigation to rings of less than 0.1-eV energy. The main purpose of the present investigation was to show the feasibility of making vortex rings so small that they can be deflected in a magnetic field.

The fact that we observed several times that vortex rings accelerated by 0.10 V needed an opposing potential of 0.11 V to stop the current completely can be understood if we assume that at least some of the rings were created, not by acceleration of thermalized ions in the electric field, but earlier, in the slowingdown of the ion pair produced by an α particle. The α particle loses 30-46 eV per ion pair (see Ref. 15), but the ionization potential of He is only 24.6 eV, so the difference must be carried away by the products of the collision: the ion and the electron. If a vortex ring is already formed by an ion in the process of being thermalized, and then is only dragged along by the applied electric field, it is quite possible that it retains some residual energy from the collision process in which the ion was created.

While using accelerating fields so small that no vortex rings were formed and the current carriers were ions only, we observed that the total current of negative ions was several times bigger than that of positive ions in the same field. This seems to indicate that the 30-46-eV α -particle losses in energy per ion pair produced are not distributed evenly between positive and negative ions. The negative charge appears to have a greater chance to escape recombination than the positive charge, at least at these temperatures, where the mean free path is long compared even with the range of the Coulomb forces.

We measured the minimum voltage between source and first grid that was necessary to produce vortex rings, and found complete confirmation of the results in Refs. 8 and 16, if the words "vortex ring" are substituted for "'runaway' particle."

If the electric field was sufficient to accelerate the ions to a drift velocity of about 30 m/sec, the first vortex rings could be observed as a current reaching the collector against a small opposing potential on the auxiliary grid. The ratio of vortex rings to ions increased sharply with increasing ion drift velocity and when, according to the universal curve of Ref. 8, the field produced an ion drift velocity of over 40 m/sec, then the current was carried practically by vortex rings alone. This region (between 30 and 40 m/sec drift velocity) corresponds to the "giant fall" observed by Careri's group,17 where the drift velocity drops from the high value of fast ions to that of vortex rings.

¹² P. E. Parks and R. J. Donnelly, Phys. Rev. Letters 16, 45 (1966).

¹³ H. London, G. R. Clarke, and E. Mendoza, Phys. Rev. 128, 1992 (1962).

¹⁴ H. E. Hall, in Proceedings of the International Conference on Superfluid Helium, St. Andrews, Scotland, 1965 (to be published).

¹⁵ F. L. Hereford and F. E. Moss, Phys. Rev. 141, 204 (1966). 16 L. Meyer and F. Reif, Phys. Rev. 123, 727 (1961). 17 G. Careri, in *Proceedings of the Ninth International Conference on Low Temperature Physics*, edited by J. G. Daunt, D. O. Edwards, F. J. Milford, and M. Yaqub (Plenum Press, Inc., New York, 1965), p. 335.

We were able to measure drift velocities of ions by measuring the Hall mobility at 0.3°K (see Ref. 7) and found velocities up to 40 m/sec, just as in Refs. 8 and 16. Huang and Olinto, 18 in their attempt to explain the steps in the ion mobility found by Careri and co-workers, 19 claimed that, at these temperatures, the step value of about 5 m/sec should be the limiting velocity for ions, since other scattering processes have become sufficiently unimportant. We did not find any evidence of a limiting or preferred drift velocity of 5 m/sec.

The values of the zero-field mobility derived from our data by using the universal curve of Ref. 8, lie within the error on the extrapolation of the curves given in Ref. 20, down to 0.5°K, and in Ref. 7 to 0.4°K. It can, therefore, be safely assumed that the scattering of the ions by phonons is approximately proportional to $T^{-3.3}$, and that the higher values proposed theoretically for the temperature dependence, $T^{-7}-T^{-9}$, cannot be reconciled with the results.21,22

The fact that impurities made it impossible to produce small vortex rings can be explained by a simple model. It has been found that even in normally pure liquid He (impurity content about 1 ppm) the light emitted from α -particle tracks shows mainly the vibrational spectra of N₂ and O₂, which probably are present in colloidal suspension, because the real solubility of O₂ of N₂ should be vanishingly small. If we assume that the charges are captured by colloidal impurities, (since we work with less than 104 charges per cm3, their number can be indeed very small) the minimum size vortex ring produced by a charged colloidal particle accelerated in the electric field is probably at least its own size. So if, for example, the colloidal particle has a diameter of 200 Å the minimum radius of a vortex ring would be 100 Å, corresponding to a minimum energy of 0.23 eV. The minimum velocity v_c necessary

for producing such a big vortex ring according to the Landau criterion,

$$v_c \geq E/p$$
, (6)

is reduced approximately proportional to (1/R) [cf. Eqs. (1) and (5) and Ref. 23] facilitating the formation of the rings. Furthermore, the colloidal particle might be bound rather strongly by the core of a vortex because it displaces regions of high fluid velocity and lowers the energy as shown by Donnelly (Ref. 24). The observation in Ref. 16, that the negative ions, which are electrons inside bubbles, do not form vortex rings in He under pressures of more than 15 atm, can be understood as the other extreme case of this model. The bubble is reduced in size by the pressure^{21,25,26} and the charge carrier becomes so small that it is more advantageous at high velocities to form a roton than a very small, and perhaps unstable, vortex ring.

The fact that we were able to purify the He by electrolysis at low temperatures (0.4-0.5°K), with electric fields such that the current carriers must have been vortex rings, indicates that any colloidal particles present must be dragged out of the drift space by vortex rings. There is also some indication that even He³ has been removed by electrolysis; it is quite possible that the He³ is attracted by the lower density in the vortex core because it can essentially reduce its zero-point energy, just as happens for electrons.27,28

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²⁷ G. Careri, W. D. McCormick, and F. Scaramuzzi, Phys. Letters 1, 61 (1962).

²⁸ B. E. Springett, D. J. Tanner, and R. J. Donnelly, Phys. Rev. Letters 14, 585 (1965).

¹⁸ K. Huang and A. Cesar Olinto, Phys. Rev. 139, A1441 (1965)

¹⁹ G. Careri, S. Cunsolo, and P. Mazzoldi, Phys. Rev. Letters 7, 151 (1961); Phys. Rev. 136, A303 (1964).

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21 I. M. Khalatnikov and V. N. Zharkov, Zh. Eksperim. i Teor.

Fiz. 32, 1108 (1957) [English transl.: Soviet Phys.—JETP 5, 905 (1957)].

22 At still lower temperatures, the mobility of ions in well

helium is limited by the scattering due to the He³ impurity, but this probably did not affect our results beyond the error of about

²³ W. F. Vinen, *Liquid Helium*, edited by G. Careri (Academic Press Inc., New York, 1963), p. 343.

²⁴ R. J. Donnelly, Phys. Rev. Letters 14, 39 (1965).

²⁵ J. Jortner, N. R. Kestner, S. A. Rice, and M. H. Cohen, J. Chem. Phys. 43, 2614 (1965).