ION MOBILITIES IN HELIUM*

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Since the early 1950's there has been a renewed interest in the study of the mobilities of ions in gases. This interest has grown in recent years with the discovery that in certain rare gases, helium and argon for example, it is possible to measure the mobility of three distinct ions under conditions where the gas purity was considered to be very good. Although various suggestions have been made to account for the presence of three ions, their identities have not as yet been established.

A brief review of some of the experimental results obtained from ion mobility measurements in helium illustrates the fact that the mobility values attributed to the molecular ion have varied considerably. In the 1930's Tyndall and his group at the University of Bristol measured the mobility of ions in helium using a glow discharge as an ion source.[1] They obtained a mobility of 20.9 cm^2/V sec (reduced to 760 Torr at 273°K) for an ion they believed to be the atomic helium ion. It was later suggested by Meyerott in 1944 that the ion involved may actually have been the molecular helium ion.[2] In 1951 Hornbeck obtained a mobility of 19 cm^2/V sec, which he attributed to the molecular helium ion, using an electron avalanche stimulated by an ultraviolet light flash as a source of ions.[3] The mobility values reported by this author lie between 17 and 20 cm^2/V sec. In 1954 Biondi and Chanin published a mobility value of 20.3 cm^2/V sec for an ion which they believed to be the molecular helium ion.[4] The ions were extracted from a high-voltage short-duration pulsed discharge.

Somewhat later, various mobility values attributed to the molecular helium ion were reported from microwave measurements of the ambipolar diffusion coefficient. These values were considerably lower than those obtained by means of the ion-transit-time method. In 1957 Oskam obtained a value of 16.3 cm^2/V sec calculated from afterglow measurements of the ambipolar diffusion coefficient.[5] Kerr and Leffel in 1961 reported ambipolar-diffusion-coefficient measurements which yielded a mobility of 16.2 cm^2/V sec.[6] An identical value was reported in 1963 by Oskam and Mittelstadt using the same measuring technique.[7]

In 1961, Patterson and Beaty[8] as well as Madson and Oskam[9] reported new ion-transit-time mobility measurements; the values obtained were 16.7 and 16.5 cm^2/V sec, respectively. Recently, Beaty and Patterson reported mobility values in helium of 10.4, 16.7, and 19.6 cm^2V sec.[10] Their measuring technique included the possibility of aging the ions before they entered the drift region. The ion with a mobility of 19.6 cm^2/V sec, however, could be observed only when sampling the ions from the plasma shortly after breakdown and then only at the lowest gas pressures used (about 3 Torr).

In spite of the variation in the published mobility values attributed to the molecular helium ion, agreement on the mobility value believed to be that of the atomic ion has been very good. The range of reported mobility values is from 10.2 to 10.8 cm^2/V sec. It appears that the ion mobilities in helium group around three different values, the first group around 10 cm^2/V sec, the second around 16 cm^2/V sec, and the third around 20 cm^2/V sec.

It has been felt for some time that it is necessary to use mass-analysis techniques combined with drift-velocity or ambipolar-diffusion-coefficient measurements in order to assign a meaningful value of mobility to a particular ion type. This technique has been employed by Saporoschenko,[11] McDaniel,[12] McAfee,[13] and Sauter, Gerber, and Oskam.[14] In our laboratory a system was constructed which also combines ion-transit-time measurements with simultaneous mass analysis. The ions are extracted from a high-voltage short-duration pulsed discharge similar to that used by Biondi and Chanin.[4] After passing through the drift region under the influence of a known uniform electric field, part of the ions enter, through a 100μ aperture, into a quadrupole mass spectrometer where they are mass analyzed. The details of the measuring system will be published elsewhere.

Preliminary studies using this measuring system indicate the presence in our helium

discharge of three helium ions having distinctly different mobilities. Figure 1 shows the mobility values of these ions as a function of E/p_0 (electric field to reduced gas-pressure ratio) at a helium pressure of 3.62 Torr. These data, although preliminary in the sense that the sensitivity of the measuring system is not as yet optimized, are representative of results obtained over a pressure range of 3 to 5 Torr.

The lower set of data refers to the atomic helium ion; these data are in very good agreement with previous results and serve to indicate that the apparatus is functioning properly. The upper two sets of data both refer to ions of mass 8. The two data sets were measured simultaneously, i.e., two ion signals appeared on the screen of the oscilloscope at the same E/p_0 value resulting in different mobility values. One set of mobility data lies in the vicinity of 16 cm^2/V sec, while the other lies in the vicinity of 20 cm^2/V sec. These values are in agreement with the two values which have previously been reported as being the mobility of the molecular helium ion. Under the present experimental conditions, ions of mass 16 were not observed, which should rule out the possibility that one of the ions appearing at mass 8 was a doubly charged oxygen ion. Although the data indicate that the mobility of the fast ion decreases with decreasing E/p_0, it should be noted that the accuracy of the measurements also decreases with decreasing E/p_0 for this type of study.

Only one ion of mass 8 was present over a wide range of discharge conditions when a narrow, low-amplitude discharge pulse was used. The corresponding mobility values were those given by the upper set of data in Fig. 1. The discharge conditions were very nearly the same as those used by Biondi and Chanin.[4] It is, therefore, believed that the ion they measured as having a mobility of 20.3 cm^2/V sec was most probably the molecular helium ion and not an impurity ion as suspected. As the discharge pulse amplitude and width were increased the second ion of mass 8 appeared. The mobility values measured for this ion lie in the vicinity of 16 cm^2/V sec.

At present it is believed that the helium ion of mass 8 having a mobility of about 20 cm^2/V sec is a newly formed helium ion, while the other helium ion of the same mass but having a mobility of about 16 cm^2/V sec is a helium ion which has been aged. This is consistent with recent results obtained during mass-spectroscopic studies of the helium afterglow.[14] In these studies it was found that for pressures above about 4 Torr, the ion with mass 8 was the dominant ion during the late afterglow period. The ambipolar diffusion coefficient measured yielded a mobility value of 16.6 cm^2/V sec.

When the experimental methods employed by the various investigators who reported mobility values of about 20 cm^2/V sec are compared, it is found that, with the exception of Tyndall, all these authors used somewhat similar methods of ion production. The feature in common appears to be that the ions studied were newly formed ions and were subjected to measurement before they could age appreciably. In contrast, the mobility values in the

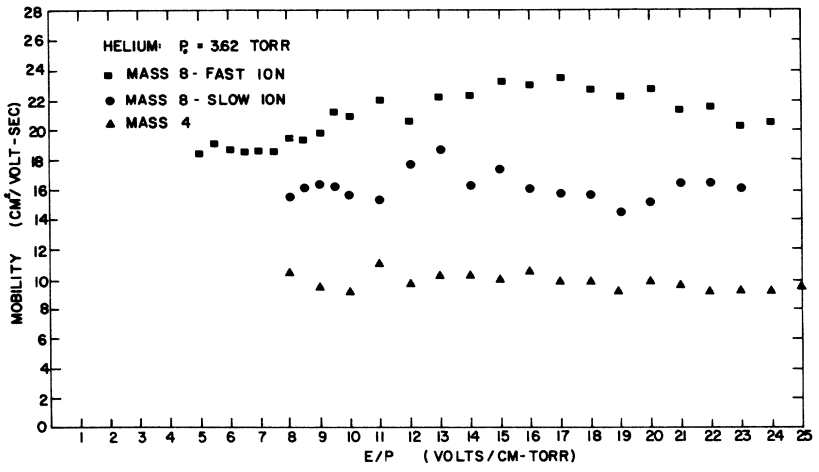

FIG. 1. Mobilities of the mass-4 and mass-8 helium ions in helium as a function of E/p_0.

vicinity of 16 cm^2/V sec were obtained during studies where the ions were considerably older. The reason that Biondi and Chanin[4] did not detect the ion having a mobility of 16 cm^2/V sec was most probably that the discharge pulses used in their studies were shorter than those at which this ion appeared in the present experiment. The value of 20.9 cm^2/V sec reported by Tyndall could probably refer to an impurity ion such as the neon ions, since it was found, during the present studies, that this ion was the dominant ion when commercial helium was not further purified by means of the cataphoretic-segregation process.[15]

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CHARACTERISTIC TEMPERATURE DEPENDENCE FOR LOW-LYING LATTICE RESONANT MODES*

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Recent investigations of impurity-induced lattice absorption in the far-infrared spectral region have demonstrated that sharp absorption lines can be associated with lattice resonant modes.[1-3] The experiments have clearly shown that a heavy impurity ion or light impurity ion coupled with weak force constants can activate a low-frequency infrared-active mode in alkali-halide crystals.

We wish to call attention to a marked temperature dependence of the resonant-mode absorption strength which has been observed. Interpreting our experimental results, we find that (1) the observed temperature dependence probably is characteristic of lattices which contain low-frequency resonant modes, and (2) a temperature-dependent contribution to the static dielectric constant should occur. Neither of these results has been previously predicted.

The experimental apparatus for the far-infrared measurements has been described elsewhere.[1] The experimental results are shown in Fig. 1 for a resonant mode absorption produced by a heavy impurity ($KI:Ag^+$) and also by a light impurity ($KBr:Li^+$). In Fig. 1(a) the absorption coefficient in the neighborhood of the resonant-mode absorption for $KI:Ag^+$ is shown for three different temperatures. As the sample temperature is increased the eigenfrequency decreases slightly and the half-width increases; however, the largest change occurs in the absorption strength ($\int A d\omega$ [cm^{-2}]). The exponential form of the temperature dependence of the absorption strength is shown in Fig. 1(b) where a semilog plot of the strength versus square of temperature can be fitted to a straight line. A similar temperature dependence is found for $KBr:Li^+$ and is shown in Fig. 1(c). Finally, the general nature of this temperature dependence is indicated by the similarity of