Determination of the Energy Level V_0 of Electrons in Liquid Argon over a Range of Densities

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The energy level V_0 of electrons injected into liquid argon was determined over the range of temperatures and densities in which the electron mobility passes through a maximum. A minimum in V_0 was found, not at the density of the mobility maximum as expected, but at a somewhat higher density.

Introduction

Electrons injected into dielectric liquids possess two measurable parameters: mobility (μ) in an electric field, and energy level (V_0) with respect to a thermal electron in the vacuum. To attempt an understanding of the state of the electron in a simple liquid, extensive measurements of μ in liquid argon were carried out over a wide range of pressure and temperature [1], revealing a spectacular rise and subsequent fall in μ as temperature was increased into the critical region. Extensive theoretical study [2—8] has not yet obtained a completely satisfactory quantitative explanation of the maximum in μ, but does suggest [2, 7] that values of V_0 and μ should be strongly connected. Measurements of V_0 in liquid argon have been carried out [9] only at a temperature near the freezing point. In the present work, V_0 has been determined for argon in the density region where the anomalous rise in μ occurs.

Experimental

V_0 was determined directly as the shift of the photoelectric wavelength threshold produced by submergence of a gold surface in liquid argon. The apparatus of Tauchert [9] was used, with the addition of a stainless steel pressure vessel to contain the gold electrode, fitted with a 1-cm thick silica window for entrance of the light. Temperature was controlled with a stream of cold nitrogen. The light from a monochromator was split, with one beam led to a photocell, current from which (i_{p}) gave a relative measure of light intensity. The remainder of the light fell on the gold surface, resulting in an emission current i_{e}. For a series of wave-lengths λ from 235—300 nm, the important data were values of the relative yield ratio $i_{\mathrm{e}}/i_{\mathrm{p}} = R$ as a function of photon energy $h\nu$.

Stability of the surface emissivity was a problem, though gold was more stable than other metals tried. Even the cleanest evaporated gold showed a slow decrease in emissivity as irradiation continued, both in vacuum and in argon, though the original emissivity was essentially recovered on standing a few minutes in the dark. The procedure adopted was to start reading at the shortest λ, increasing λ in 5-nm steps until beyond the emissivity threshold, then to decrease λ again in 5-nm steps, and average the two values given for each λ. This procedure gave reproducible results.

It had been the practice [9, 10] to fit observations to Fowler's emissivity function [11], but systematic deviations always were found between the data and the function. We therefore evaluated the data more directly by a simple graphical comparison: log R was plotted vs. $h\nu$ on transparent graph paper, for a vacuum run and an argon run made at a similar temperature on the same day, and the curves were made to coincide by vertical and horizontal sliding of the paper. The horizontal shift gives V_0. The method implies that the percentage of photoelectrons which succeed in escaping into the argon is independent of $h\nu$. This was true in all previous work with hydrocarbons [10] and with argon at 85 K [9]. Figure 1(a) shows that it holds also at 121 K, which is just below the range where μ shows its sharp rise with increasing temperature. At higher temperatures (Fig. 1b) the vacuum and argon curves cannot be made to coincide for the higher

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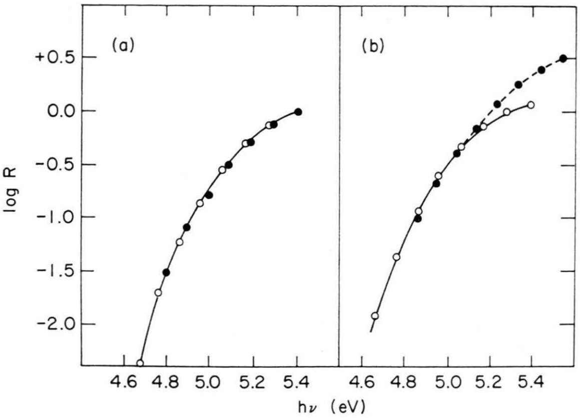

Fig. 1. Fitting curves of log photoelectric current (arbitrary units) vs. photon energy; ○, vacuum, ●, liquid Ar. (a) 121 K, 41 atm., $n = 17.8 \times 10^{21}$ cm^{-3}; Ar curve moved 0.13 eV to the right and 2.20 log units upward. (b) 151 K, 66 atm., $n = 12.9 \times 10^{21}$ cm^{-3}; Ar curve moved 0.27 eV to the right and 2.56 log units upward.

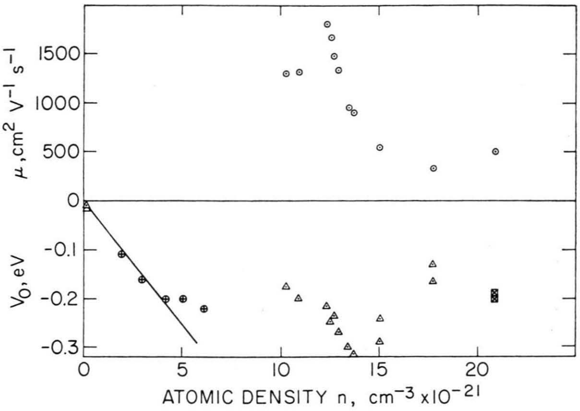

Fig. 2. Lower half, V_0 values for gaseous and liquid argon: △, present work; ⊠, Ref. [9]; ⊕, Ref. [12]. Upper half, mobility values from Ref. [1] at the temperatures and pressures of the runs plotted in the lower half.

$h\nu$ values, so here the escaping fraction increases with initial energy. Here V_0 was obtained from the shift which gave the best fit in the threshold region. Our V_0 values appear reproducible to about ± 0.03 V.

Results and Discussion

Table 1 summarizes our results. The V_0 values are graphed in the lower part of Fig. 2 as a function of argon density, together with Tauchert's values [9] at a higher density and values obtained by a spectroscopic method [12] in argon gas. The line drawn from the origin shows the values of V_0 expected from the shift of 10 cm^{-1} reported [13] for ionization potentials of alkali metal atoms in argon at 1 atm. pressure. Our results in argon gas at 5.3 atm. and 298 K are in fair agreement. A run at 148 atm. and 298 K could not be used because the curve of log R vs. $h\nu$ was so different from the vacuum curve that no good estimate of V_0 was possible.

Table 1. Summary of V_0 Data.

Run No.	Pressure, atm.	T[K]	Atomic density[a] n, cm^{-3} $\times 10^{-21}$	Shifts, Ar to vac.	
				$\Delta h\nu = -V_0$	Δ log R
11–6	5.28	298	0.13	0.015	0.76
11–7	5.27	298	0.13	0.010	0.82
11–29	41	121	18	0.130	2.20
11–30	39	121	18	0.165	2.18
12–12	64	148	13.5	0.30	2.04
12–13	75	150	13.7	0.315	2.36
12–14	66	151	12.9	0.27	2.56
12–15	72	153	12.7	0.235	2.60
12–16	77	165	12.6	0.245	2.66
12–17	87	158	12.3	0.215	2.56
12–22	83	145	15.1	0.285	2.10
12–23	82	144	15.1	0.24	2.08
12–24	65	155	10.9	0.20	1.94
12–25	71	159	10.3	0.175	1.92

[a] Calculated from data in U. S. Bureau of Standards Report NSRDS-NBS-27.

An approximate theory predicts that a maximum in μ should correspond to a minimum in V_0 occurring at the same density, and this was shown [14] to be true for the liquids neopentane and tetramethylsilane. But in argon, where the maximum in μ occurs at atomic density $n = 1.20 \times 10^{22}$ cm^{-3} for a wide range of pressures, the minimum in V_0 occurs at a distinctly higher density, with n about 1.4×10^{22} cm^{-3}. In a more elaborate theory [7], μ is shown to depend not only on V_0 and its first derivative with respect to n, but also on the second and third derivatives. In the two liquids mentioned above, the curves of V_0 vs. n were shown to be quite flat and apparently symmetrical about the minimum over a range in n of a factor of 2.5, so the second derivative is small and essentially constant. In argon, the minimum in V_0 is narrower and the second and apparently the third derivatives are more important, so the deviation in n between maximum μ and minimum V_0 is not too surprising. An adequate theory for the absolute values of V_0 in argon and other liquids, and for the details of its variation with n, is not available at present.

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