

1 **Electron thermalization length in solid para-hydrogen at low-temperature**

2 A. F. Borghesani,^{1, a)} G. Carugno,² G. Messineo,³ and J. Pazzini⁴

3 ¹⁾*CNISM unit, Department of Physics & Astronomy, Università degli Studi di Padova*
4 *and Istituto Nazionale Fisica Nucleare, sez. Padova, Italy*

5 ²⁾*Istituto Nazionale Fisica Nucleare, sez. Padova, Italy*

6 ³⁾*Istituto Nazionale Fisica Nucleare, sez. Ferrara, Italy*

7 ⁴⁾*Department of Physics & Astronomy, Department of Industrial Engineering,*
8 *and Department of Information Engineering, Università degli Studi di Padova and*
9 *Istituto Nazionale Fisica Nucleare, sez. Padova, Italy*

10 (Dated: 1 August 2023)

11 We report the first ever measurements of the thermalization length of low-energy elec-
12 trons injected into solid para-hydrogen at a temperature $T \approx 2.8$ K. The use of the pulsed
13 Townsend photoinjection technique has allowed us to investigate the behavior of quasi-free
14 electrons rather than of massive, slow negative charges as reported in all previous litera-
15 ture. We have found an average thermalization length $\langle z_0 \rangle = 26.1$ nm, which is 3 to 5 times
16 longer than that in liquid helium at the same temperature.

^{a)}Electronic mail: armandofrancesco.borghesani@unipd.it

17 I. INTRODUCTION

18 An active field of research is the measure of the permanent electric dipole moment of the
19 electron to look for CP violation phenomena beyond those included in the standard model of
20 particle physics¹⁻³. Barium monofluoride (BaF) is a promising polar molecule to be used for this
21 goal because its outermost electron in the HOMO is acted upon by an intrinsic electric field whose
22 strength is otherwise unattainable in laboratory⁴.

23 The PHYDES collaboration aims at embedding BaF molecules in a solid matrix of para-
24 hydrogen (p-H₂) at cryogenic temperatures or in other cryogenic noble gas solid environments.
25 This matrix isolation technique limits the host-guest interaction, enhances the density of the em-
26 bedded molecules, and reduces their diffusion prowess^{5,6}.

27 Solid para-hydrogen at low temperature is a matrix of great interest because of its remarkable
28 properties due to its quantum nature. The antiparallelism of the nuclear spins leads to a config-
29 uration of lowest energy. The solid crystallizes in hexagonal closed-packed structure (*hcp*), the
30 average nearest-neighbor distance is ≈ 0.38 nm with a molecular mean-squared displacement of
31 ≈ 0.048 nm⁷⁻¹⁰. Its number density at $T = 4.2$ K is fairly high, $N \approx 2.6 \times 10^{28}$ m⁻³, and the solid
32 can be considered to consist of an assembly of molecules all translationally localized at lattice
33 sites but freely rotating even in the zero-temperature limit.

34 Embedding neutral BaF molecules in the solid p-H₂ matrix is yet an unsolved technical prob-
35 lem. Actually, solid BaF₂ is evaporated in a glow discharge chamber. The BaF₂ molecules are
36 dissociated and ionized. BaF⁺ cations are then extracted by means of electrostatic lenses and
37 mass-selected by a Wien filter. Eventually, the cations are electrostatically slowed down to a final
38 kinetic energy of ≈ 5 eV and are admitted into the low-temperature chamber where they mix with
39 the p-H₂ gas that has to be condensed to form the solid. Details on this technique will appear in a
40 future publication.

41 We plan to investigate the possibility of neutralizing the cations embedded in the solid by
42 injecting into it electrons from a photocathode irradiated with a short UV laser pulse. To achieve
43 this goal the knowledge of the energetics and dynamics of low-energy electrons in the solid p-H₂
44 matrix is of paramount importance. However, to the best of our knowledge, there are only a couple
45 of papers about excess negative charge carriers in solid p-H₂^{11,12} and a few more in solid^{13,14} and
46 liquid H₂¹⁵⁻¹⁸.

47 Almost all the experimental papers found in literature are dealing with negative charges of

48 unspecified nature. In those experiments ionization is obtained by either using an energetic elec-
 49 tron beam of $\approx 45 \text{ keV}^{13}$ or β -rays from Tritium sources whose average energy is in excess of
 50 $5 \text{ keV}^{11,12,14-16}$ whereas the dissociation energy of H_2 is $\approx 4.5 \text{ eV}$. These energy values are suffi-
 51 cient for both ionization and repeated dissociation events of the hydrogen molecules that may lead
 52 to the production of stable H^- ions or even $(\text{H}_2)_n^-$ cluster ions.

53 The analysis of the phenomenology of the drift mobility measurements of the negative excess
 54 charges in both liquid and solid hydrogen and in solid p- H_2 convincingly demonstrates that differ-
 55 ent ionic species were detected rather than quasi-free electrons.

56 In liquid H_2 in the range $12 \text{ K} < T < 34 \text{ K}$, the value of the drift mobility μ of negative species is
 57 comprised in the range $1.5 \times 10^{-2} \text{ cm}^2/\text{Vs} < \mu < 3 \times 10^{-2} \text{ cm}^2/\text{Vs}$, comparable to the value in liq-
 58 uid helium¹⁹, and shows evidence of the presence of two different negatively charged species^{15,20}.
 59 This behavior is rationalized by assuming that, owing to the very large and positive ground-state
 60 energy of an electron in the conduction band of the liquid $V_0 \approx 1.9 \text{ eV}^{17}$ and because of the strong,
 61 repulsive, short-range exchange forces both electron bubbles (i.e., electrons self-localized in empty
 62 cavities) and ionic bubbles²¹ (i.e., H^- ions or $(\text{H}_2)_n^-$ cluster ions surrounded by a void shell) both
 63 coexist in the liquid¹⁸.

64 In solid hydrogen the mobility of the negative charges is very low, roughly ranging from
 65 $4 \times 10^{-7} \text{ cm}^2/\text{Vs}$ to $8 \times 10^{-6} \text{ cm}^2/\text{Vs}$ in the range $12 \text{ K} < T < 14 \text{ K}^{13,14}$, also depending on the
 66 applied drift field, and strongly decreases with decreasing temperature. In order to rationalize
 67 these results several hypotheses have been suggested. The first one is that electrons deform the
 68 not-so-rigid solid (its surface tension is estimated to be $\sigma \approx 4.9 \times 10^{-3} \text{ J/m}^2$ at $T = 2.8 \text{ K}^{22}$) and
 69 self-localize in empty bubbles giving origin to electron bubbles as was suggested for the case of
 70 solid helium²³, in which the mobility values are of comparable magnitude²⁴⁻²⁶. The second one is
 71 that the charge carriers are localized by the formation of small polarons and their transport occurs
 72 via phonon-assisted- or hopping processes^{13,27}. Whatever the explanation might be, it is clear
 73 from the experiments that the charge carriers cannot be quasi-free electrons.

74 Finally, in solid p- H_2 the drift mobility of excess negative charge carriers turns out to be
 75 extremely low. In the temperature range $13.6 \text{ K} > T > 5.8 \text{ K}$ μ decreases from a value $\mu \approx$
 76 $6 \times 10^{-6} \text{ cm}^2/\text{Vs}$ down to $\mu \approx 2 \times 10^{-11} \text{ cm}^2/\text{Vs}^{11,12}$. Above $T = 10 \text{ K}$ the diffusion coefficient D
 77 of negative charges, obtained from the mobility by exploiting the Nernst-Debye-Einstein relation-
 78 ship $D/\mu = k_B T/e$, in which k_B is the Boltzmann constant and e is the electron charge, closely
 79 resembles the diffusion coefficient of ortho-hydrogen molecules and is therefore rationalized by

80 assuming a mechanism of thermally activated diffusion of charged point defects interacting with
 81 vacancies²⁸. Below $T = 8$ K also a mechanism of quantum tunneling of vacancies has to be intro-
 82 duced. In this way the activation energy and the free energy for vacancy formation are determined
 83 and are in good agreement with the theoretical estimate²⁹ and with their determination in NMR
 84 experiments³⁰. Moreover, the authors warn that some differences in the experimental outcome are
 85 to be attributed to the anisotropic crystal structure¹¹.

86 The nature of the negative charge carriers has been identified with localized point charge defects
 87 that interact with the vacancies in the crystal¹¹. It is very plausible that such entities are massive
 88 H^- anions or the even more massive $(\text{H}_2)_n^-$ cluster ions as possible products of the technique used
 89 to inject charges in the solid. Moreover, the presence of H_2^- anions has been observed in γ -ray
 90 irradiated, pure solid p- H_2 in high-resolution ESR experiments³¹, whereas electron bubbles are
 91 produced in place of H_2^- anions only if the solid p- H_2 contains a small amount of deuterium³².
 92 Actually, similar mechanisms of interaction between charge carriers and vacancies have been ob-
 93 served in solid deuterium, in which negative charges are assumed to be electron bubbles³³.

94 The whole wealth of experimental results and their interpretation strongly indicates that until
 95 now the behaviour of quasi-free electrons in fluid and solid hydrogen, including solid p- H_2 , has
 96 not been investigated yet. Therefore, we have carried out an experiment in which bunches of
 97 low-energy electrons are photoinjected into the solid by means of short pulses of a UV laser. We
 98 report in this paper the results of the measurements of the thermalization length of such quasi-free
 99 electrons in solid p- H_2 at $T \approx 2.8$ K.

100 The paper is organized as follows. In Sect. II details on the experimental setup and procedure
 101 are given. In sect. III we present and discuss the experimental results. Finally, some conclusions
 102 are drawn in Sect. IV.

103 II. EXPERIMENTAL DETAILS

104 A thorough description of the whole apparatus will appear in a subsequent paper. Here we only
 105 give a brief and schematic description of the experimental setup and technique.

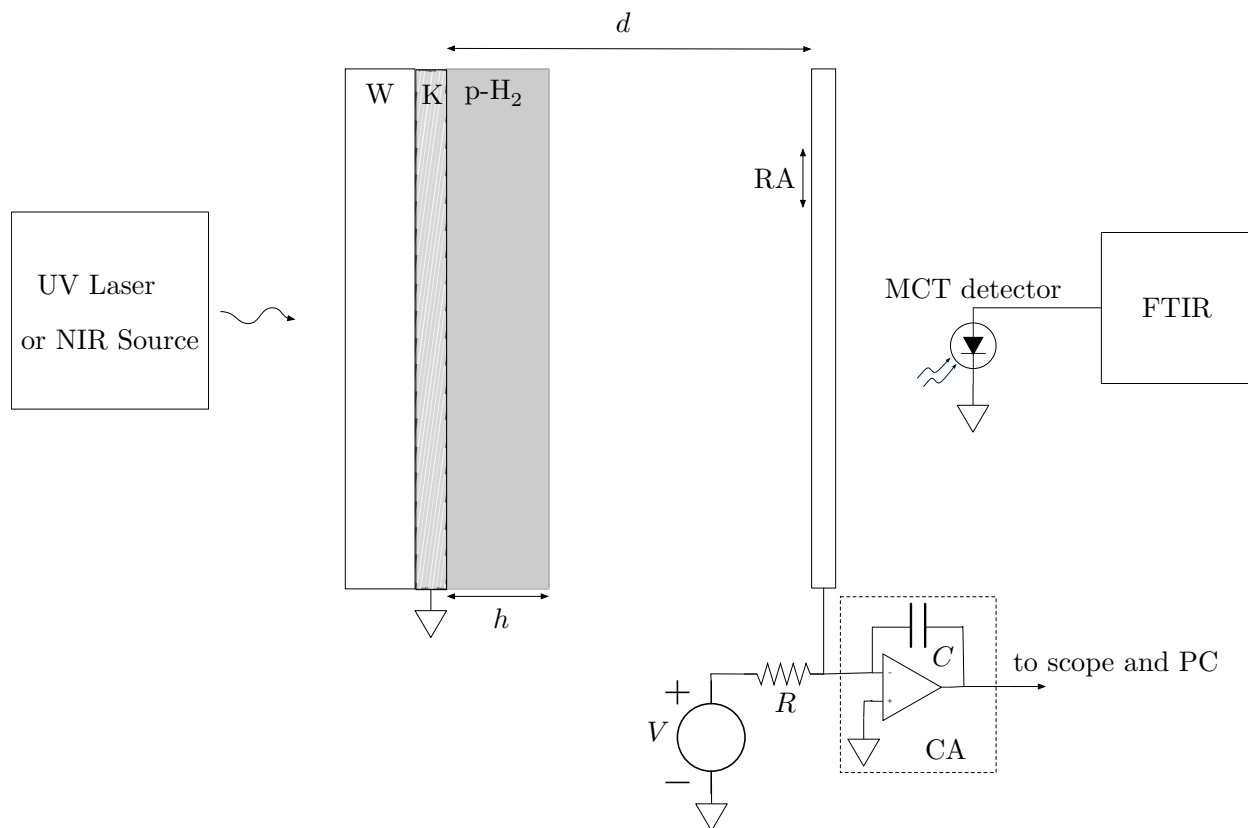
106 The measuring cell is located in a cryostat and is cooled down to $T \approx 2.8$ K using a commer-
 107 cially available cryocooler. The cell can be optically accessed through suitable viewports in the
 108 cryostat and is evacuated down to a residual pressure $P_{\text{res}} \approx 10^{-6}$ Pa.

109 Normal hydrogen gas is enriched in the para-hydrogen fraction by exploiting standard tech-

Electron thermalization length in solid para-hydrogen at low-temperature

110 niques^{34–36}. The gas is precooled to $T \approx 77$ K by slowly flowing it in a capillary immersed in a
 111 liquid N₂ bath. Subsequently, the gas is flown through a copper tube, kept at $T \approx 22$ K by means
 112 of an ancillary cryocooler. The tube is packed with Fe₂O₃ fine powder that acts as a catalyst for
 113 the ortho→para conversion. The gas flow rate is controlled by a leak valve and measured by a
 114 flow meter. The gas enriched in the para fraction is then admitted into the measuring cell at low
 115 pressure. During the growth of the solid p-H₂ film the gas pressure is about $P \approx 7 \times 10^{-5}$ Pa.

116 A set of windows and electrodes is located in the cell to allow us to monitor the growth and
 117 purity of the p-H₂ film and to inject electrons in the solid film and collect them. A conceptual,
 118 simplified, schematics of the electrodes and windows assembly is shown in Figure 1 A sapphire



119 FIG. 1. Conceptual scheme of the setup for measuring both the thickness of the p-H₂ solid film during its
 120 growth and the amount of photoinjected charges after the film growth is stopped. Symbol meaning: W =
 121 optical substrate. K = 10 nm thin gold cathode, p-H₂ = para-hydrogen film of thickness h . RA = retractable
 anode. d = drift distance. V = d.c. power supply. C = integrating capacitor. R = high-impedance resistor.
 CA = charge amplifier. FTIR = Fourier-Transform InfraRed interferometer.

122 contact with it. The thin Au film is grounded and acts as the photocathode in a diode configuration
 123 of the electrodes.

124 At regular time intervals the thickness of the growing p-H₂ film is monitored by recording
 125 its near-infrared (NIR) absorption spectrum, as described in literature^{37,38}. The radiation of the
 126 near-infrared source (Thorlabs, model SLS202L/M) impinges on the back of the window, passes
 127 through the p-H₂ film and is detected by a MCT detector (Hamamatsu, mod. P7163) whose output
 128 is amplified and fed to the FTIR interferometer (Bruker, mod. Equinox 55), thereby yielding the
 129 absorption spectrum $I(\tilde{\nu})$ as a function of the wavenumber $\tilde{\nu}$ in cm⁻¹.

130 According to literature the film thickness h and the ortho-H₂ fraction f can be computed from
 131 the optical density $OD = -\log[I(\tilde{\nu})/I_0(\tilde{\nu})]$, in which I_0 is the spectrum recorded in absence of
 132 p-H₂, respectively as^{37,38}

$$133 \quad h(\text{mm}) = 0.048 \times \int_{4495}^{4525} OD(\tilde{\nu}) d\tilde{\nu}, \quad (1)$$

$$134 \quad f = \frac{0.124}{h} \times \int_{4151}^{4154} OD(\tilde{\nu}) d\tilde{\nu}, \quad \text{or} \quad f = \frac{0.0787}{h} \times \int_{4732}^{4742} OD(\tilde{\nu}) d\tilde{\nu} \quad (2)$$

135 A typical OD spectrum recorded after ≈ 3 hrs condensation is reported in Figure 2. In all ex-
 136 perimental runs we obtained a constant growth rate of $\approx 100 \mu\text{m/hr}$ and a constant ortho fraction
 137 $2\% < f < 3\%$.

138 After the solid film thickness has reached a value between 500 and 600 μm the gas inlet is
 139 closed and the film growth is almost stopped, its growth rate dropping down to $\approx 0.5 \mu\text{m/hr}$,
 140 whereas the ortho-hydrogen content remains constant for a longer time interval after the photoin-
 141 jection experiment has been completed.

142 Once the solid film has reached the desired thickness, the retractable anode is set in position
 143 in front of the cathode by means of a suitable manipulator. The NIR source is replaced by a 266-
 144 nm pulsed laser. The laser, whose spot has a diameter $\phi \approx 8$ mm, impinges on the back of the
 145 Au cathode and photoextracts electrons. The laser pulse has a duration $\Delta t \approx 10$ ns and an energy
 146 per pulse that can be varied in the range $50 \mu\text{J} < W < 150 \mu\text{J}$, but is kept constant during the
 147 measurement. The laser pulse repetition rate is set at 10 Hz.

148 A d.c. power supply energizes the drift capacitor up to 1 kV and sets the desired value of the
 149 electric field in the drift region of length $d = 1$ cm. The current induced in the circuit by the drifting
 150 photoextracted electrons is integrated by a charge amplifier (i.e., an active integrator) whose time

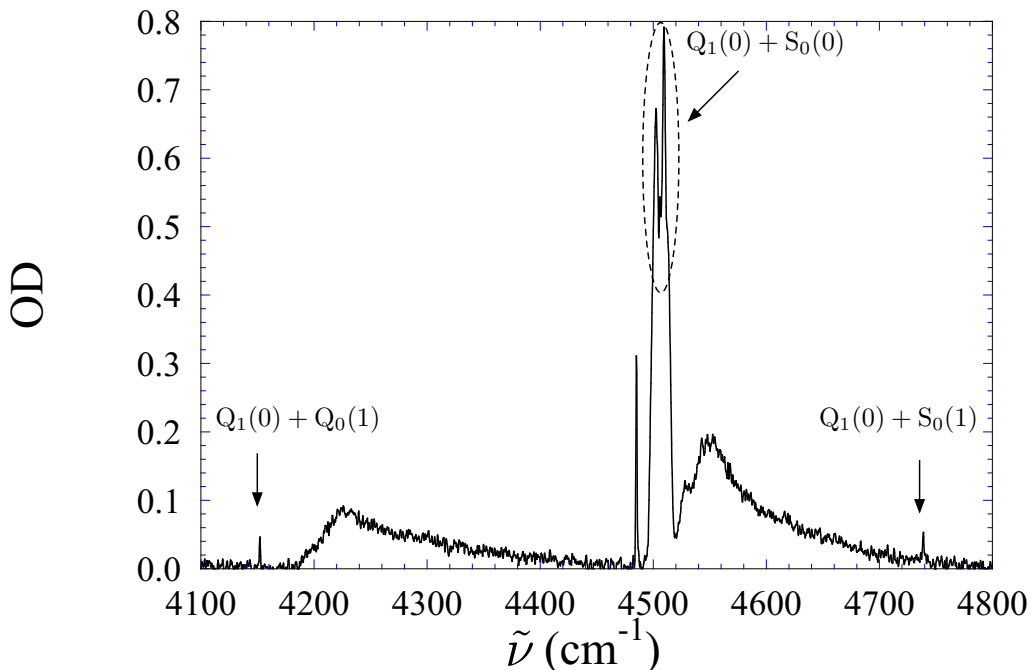


FIG. 2. Optical density spectrum of the solid p-H₂ film after 3 hrs condensation. The ro-vibrational transitions used for computing the film thickness are enclosed in the dashed ellipse. The lateral bands are analyzed to obtain the ortho-H₂ concentration. The meaning of the notation of the ro-vibrational transitions can be found in literature³⁶.

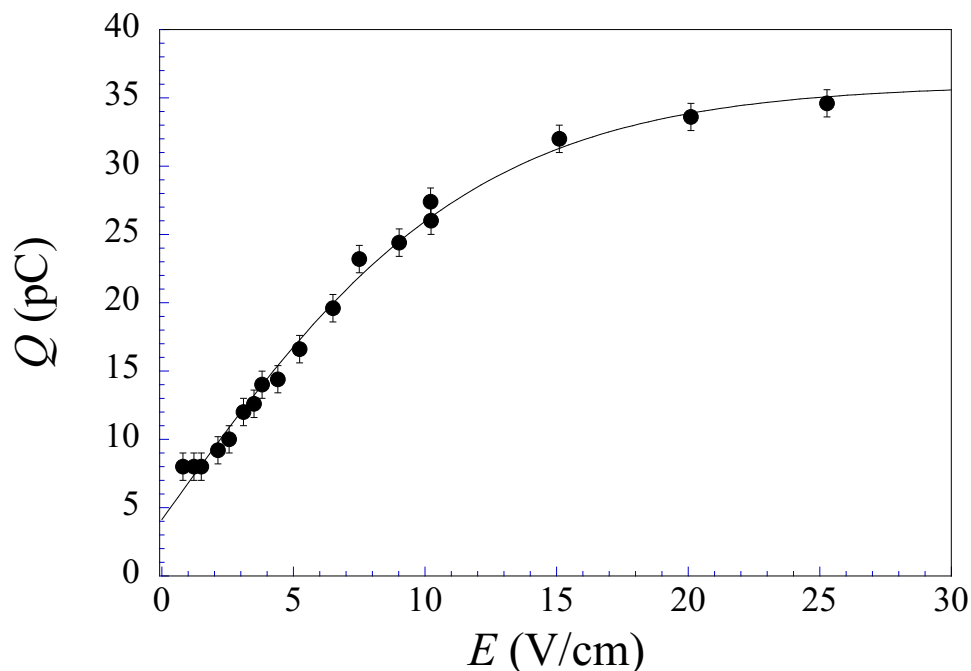
151 constant is $\tau \approx 400 \mu\text{s}$ and whose output amounts to $\approx 0.25 \text{ mV/fC}$. The charge amplifier output is
 152 connected to a digital scope. For each setting of the drift field, 2000 signals are averaged together
 153 in order to improve the signal-to-noise ratio. The averaged signals are then fetched by a PC for
 154 offline analysis.

155 III. EXPERIMENTAL RESULTS AND DISCUSSION

156 In this Section we report the experimental results and their rationalization.

157 **A. Characterization of photoextraction in vacuo**

158 First of all, the behavior of the system has to be characterized *in vacuo*. We report in Fig-
 159 ure 3 the charge photoextracted under vacuum conditions. The amount of photoextracted charge



160 FIG. 3. Electric field dependence of the charge collected at the anode *in vacuo*. The solid is only a guide
 for the eye.

161

162 increases with increasing field, as expected, and saturates for an electric field strength as weak
 163 as $E \approx 15$ V/cm, when practically the whole amount of electrons produced by the laser pulse is
 164 collected.

165

166 The energy distribution of the photoelectrons is not known, as is the work function of the
 deposited Au film. We directly measured the highest energy of the electrons by reversing the
 167 electric field polarity in order to nullify the collected charge, thereby obtaining $\epsilon_{\max} = (0.56 \pm$
 168 $0.05)$ eV. The amount of the collected charge proportionally varies with the laser pulse energy, its
 169 electric field dependence remaining the same.

170 B. Measurements with solid p-H₂ in place

171 The measurements of photoextraction have been carried out after the thickness h of the solid p-
172 H₂ film reached a value between 500 μm and 600 μm . If the applied potential difference between
173 anode and cathode is V , the electric field strength E in the dielectric film is given by

$$174 \quad E \text{ (V/cm)} = \frac{V}{K(d-h) + h} \approx 0.792V, \quad (3)$$

175 in which V is measured in Volt, $d = 1 \text{ cm}$ is the drift distance and $K \approx 1.28$ is the relative dielectric
176 constant of solid p-H₂^{39,40}. An accurate determination of h is not extremely important as a 20%
177 uncertainty in h brings about a negligible 0.2% uncertainty in E . The electric field strength in
178 the p-H₂ vapor is $E_v \text{ (V/cm)} = KE \approx 1.01V \text{ (V)}$. We have investigated the range $10 \text{ V/cm} \lesssim E \lesssim$
179 600 V/cm .

180 1. Signal waveform analysis

181 Some preliminary pieces of information can be gathered by inspecting the acquired signal
182 waveforms, such as the one shown in Figure 4. First of all, we note that the amount of collected
183 charge is a factor more than 2000 smaller than that collected *in vacuo* with a much smaller electric
184 field strength. We will discuss this reduction later in greater detail. For now, we want to stress the
185 fact that the signal rise time $\Delta t \simeq 0.7 \mu\text{s}$ is independent of the applied electric field and is solely
186 due to the response of the electronics. If electrons injected into the solid were prone to dissociate
187 H₂ to form H⁻ anions or to form electron bubbles, their drift time through the solid would be no
188 shorter than several tens of seconds. However, we do not observe any charge pile-up in the solid
189 in spite of the quite high laser pulse repetition rate.

191 On the other hand, the transit time of electrons emerging into the vapor is estimated to be
192 several orders of magnitude smaller than the rise time of our electronics because of the extremely
193 low density of the vapor at $T \approx 3 \text{ K}$. Unfortunately, there are no measurements of electron drift
194 velocity v_D in p-H₂ gas for so high an electric field strength and at so low a temperature except
195 those at $T = 76.8 \text{ K}$, at a lowest pressure $P \sim 500 \text{ Pa}$, and at the highest reduced field $E/N \sim 2.6 \text{ Td}$
196 ($1 \text{ Td} = 10^{-21} \text{ V m}^2$), yielding a drift velocity $v_D \approx 3.3 \times 10^6 \text{ cm/s}$ ^{41,42}. Just to carry out a back-of-
197 the-envelope estimate, we assume that the pressure in our experiment is so low that the vapor can
198 be treated as an ideal gas because the equation of state of p-H₂ is only known for $T > 13.8 \text{ K}$ ⁴³.
199 We, thus, get a gas number density $N \approx 2 \times 10^{18} \text{ m}^{-3}$, thereby yielding $E/N \approx 5 \times 10^5 \text{ Td}$ for $E =$

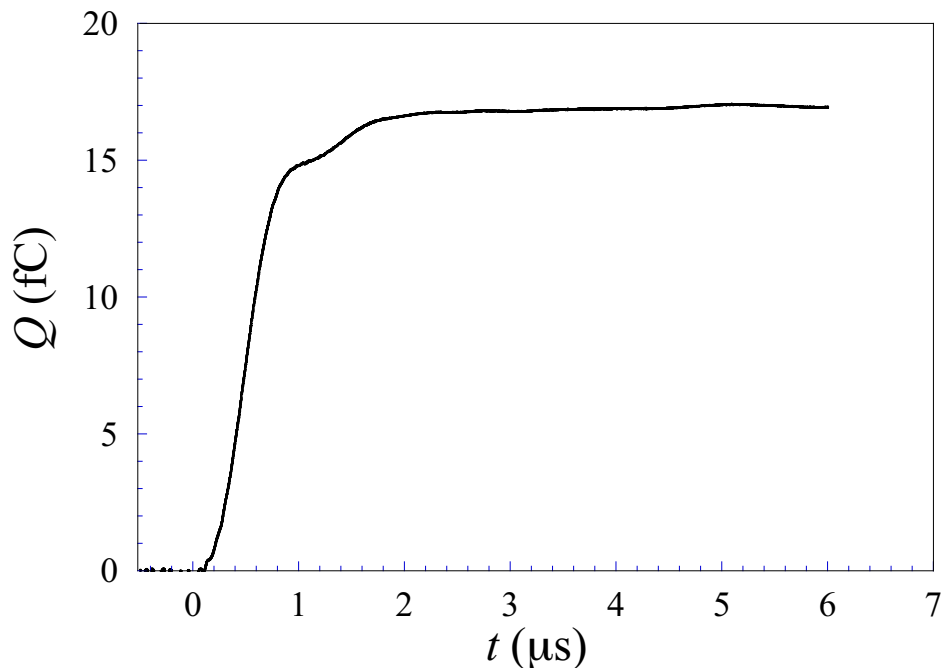


FIG. 4. Time evolution of the collected charge in presence of the solid p-H₂ film recorded with $E = 280$ V/cm.

200 10 V/cm. As there are no hints at how fast electrons travel at such huge a reduced field strength, we
 201 just assume that they move *in vacuo* and obtain a maximum final speed $v \sim 2 \times 10^8$ cm/s, thereby
 202 leading to a transit time $\tau \sim 5$ ns, much shorter than the rise time of the electronics.

203 This analysis of the waveform leads to the conclusions that: i) electrons are actually injected
 204 into the solid in which they move at high speed, and ii) neither low mobility electron bubbles nor
 205 anions form.

206 2. *Electric field dependence of the photoextracted charge*

207 The typical outcome of the experiment is shown in Figure 5, in which the amount of the charge
 208 collected at the anode is plotted as a function of the electric field E in the p-H₂ film.

210 As previously noted, the electrons photoinjected into the solid p-H₂ film are epithermal. Upon
 211 injection electrons will lose their excess energy by collisions in the solid until they get thermalized

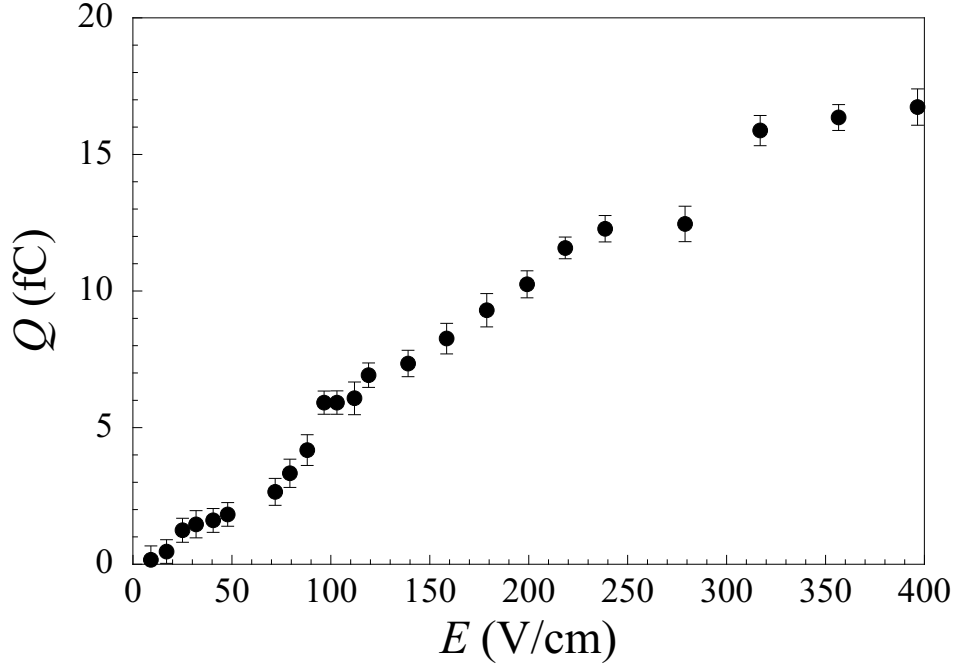


FIG. 5. Collected charge Q vs the electric field E acting on the p-H₂ film.

212 and then propagate in the conduction band of the crystal. The model used to describe the injection
 213 process has been thoroughly described in literature⁴⁴⁻⁴⁶. We briefly recall here its main features.

214 The electrons injected in the solid film close to the Au/p-H₂ interface are under the influence
 215 of their image force, which acts so as to push them back to the cathode, and of the externally
 216 applied electric field E , which pulls them towards the collector. The potential energy (in eV) of
 217 the electrons is given by

$$218 \quad \varphi(z) = V_0 - Az^{-1} - Ez \quad \text{for } z \geq 0. \quad (4)$$

219 Here, z is a coordinate orthogonal to the Au/p-H₂ interface where the coordinate origin is located,
 220 and V_0 is the electron ground state energy at the minimum of the conduction band in the solid. The
 221 coefficient A takes into account the dielectric properties of the solid film and is given by⁴⁷

$$222 \quad A = \frac{e}{16\pi\epsilon_0 K} \left(\frac{K-1}{K+1} \right) \approx 0.345 \text{ eV } \text{\AA}. \quad (5)$$

223 $\varphi(z)$ has a maximum at $z_m = \sqrt{A/E}$ where the Schottky barrier lowering (in eV) amounts to

224 $\Delta\phi = -2Ez_m$. In our experimental conditions $20\text{ nm} \lesssim z_m \lesssim 200\text{ nm}$, whereas $4 \times 10^{-5}\text{ eV} \lesssim$
 225 $|\Delta\phi| \lesssim 3 \times 10^{-4}\text{ eV}$ is negligible.

226 We note that the distance of the potential maximum from the Au/p-H₂ interface is always much
 227 shorter than the solid film thickness, $z_m \ll h$. However, we cannot be sure that z_m is greater or
 228 comparable to the electron mean free path (mfp) ℓ in the solid. We can reasonably assume that
 229 this is the case. Actually, on one hand, if we could make the hypothesis that the molecules in the
 230 solid were a collection of independent scattering centers, we would get $\ell \approx 0.4\text{ nm}$ by using the
 231 recommended value for the e-H₂ scattering cross section at low energy ($\sigma_s \approx 9.2 \times 10^{-20}\text{ m}^2$)⁴⁸.
 232 This mfp value, surprisingly close to the nearest-neighbor distance in the solid, is far too short to be
 233 reasonable. Actually, at low energies where the electron wavelength is comparable or even larger
 234 than the typical intermolecular distance, we expect deep modifications of the gas-phase behavior.
 235 Coherent scattering of the electron wave function within the solid may even deeply lower the gas-
 236 phase cross section⁴⁹. On the other hand, a mfp values of a few tens of times larger than the
 237 nearest-neighbor distance might be more realistic. In any case, we assume that $z_m \gtrsim \ell$.

238 A first attenuation of the amount of charge injected in the solid with respect to the vacuum case
 239 is due to the quantum reflection of the electron wavefunction at the barrier V_0 . This phenomenon
 240 strongly reduces the amount of charge that can be collected at the anode, as experimentally ob-
 241 served.

242 Once successfully injected into the solid, an electron loses its excess energy by scattering off
 243 molecules, or phonons, or structural defects. Thus, electrons undergo a random walk through the
 244 medium with the possibility that somewhere thermalization occurs due to some inelastic process
 245 (as, for instance, by inducing rotational excitation in the H₂ molecules). Several possible paths
 246 for electrons can be singled out. The first possibility is that the electron is backscattered towards
 247 the cathode at its first collision⁵⁰ or may backdiffuse towards the cathode before thermalization
 248 occurs. These two processes do not depend on the position of the potential maximum. A second
 249 possibility is that the electron thermalizes for $z < z_m$ and backdiffuses towards the cathode. All
 250 these processes lead to a reduction of the amount of charge that can be collected at the anode. The
 251 third and final possibility is that thermalization occurs for $z > z_m$ and the electron is collected at
 252 the anode because of the pull exerted by the external field. In some sense, the potential maximum
 253 acts on the electrons as a gate at a distance z_m from the cathode.

254 By taking into account all the aforementioned effects, it has been shown that, at low temper-
 255 ature and high density, the amount of charge injected into the solid and collected at the anode is

256 accurately described by the following equation^{44,45,51}

$$257 \quad \frac{Q}{Q_0} = \frac{B}{D} e^{-z_m/z_0}, \quad (6)$$

258 in which Q_0 is the amount of charge that would be collected *in vacuo*, and z_0 is the thermalization
 259 length. B is a coefficient that describes the effect of the barrier and could be computed if the energy
 260 distribution function of the photoelectrons were known^{52,53}. In our case, B is constant because the
 261 energy distribution function of the photoextracted electrons does not change from run to run. D is
 262 a term that describes the backdiffusion process. It can be written as $D = 1 + Cz_0/\ell$, where C is a
 263 numerical constant, and does not depend on z_m . Moreover, as all the measurements with the solid
 264 film in place have been carried out for $E \gtrsim 15 \text{ V/cm}$, Q_0 can also be considered as a constant.

265 In Figure 6 we report the experimental results for the collected charge as a function of the

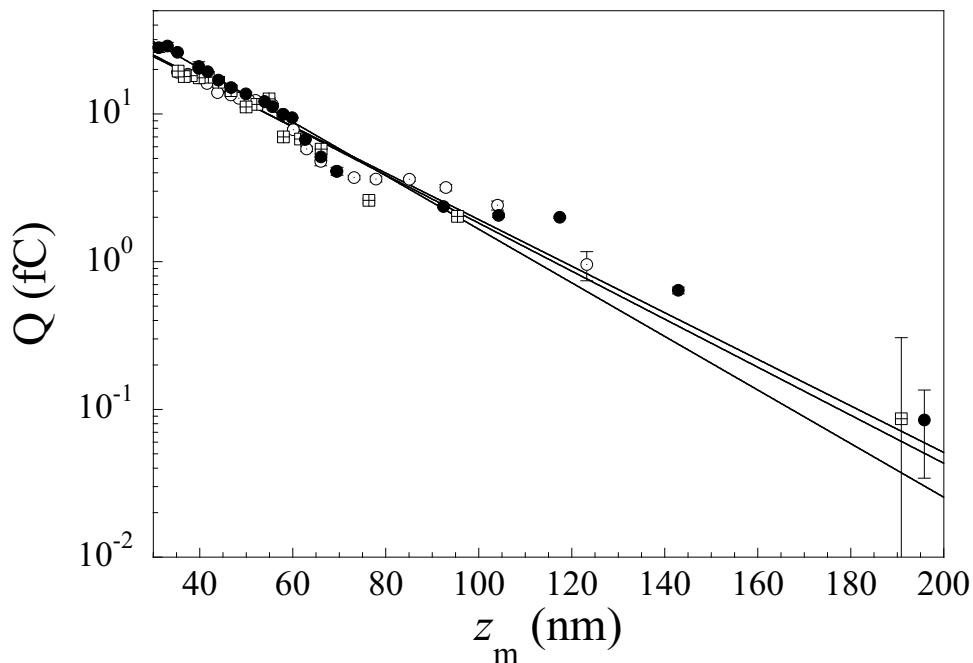


FIG. 6. The charge collected at the anode Q as a function of the coordinate z_m of the potential energy maximum measured in the three experimental runs. The charge in each run has been normalized to the laser energy. The solid lines are the best fit curves based on Eq. (6).

266

267

268 coordinate z_m of the potential energy maximum obtained in the three different runs we have carried
 269 out at roughly one month interval from each other. The charge amount in each run has been
 270 normalized to the laser energy.

271 The experimental data satisfactorily follow the exponential law Eq. 6. We obtain three indepen-
 272 dent determinations of the thermalization length that are reported in Table I. The three different

run	z_0 (nm)	uncertainty (nm)
1	25.6	2.0
2	29.1	2.8
3	25.7	1.2

TABLE I. Thermalization length determinations in the three experimental runs with the relative uncertain-
 ties.

273

274

275 determinations of the thermalization length are in good agreement with each other, thereby yield-
 276 ing a weighted average

277

$$\langle z_0 \rangle = (26.1 \pm 1.0) \text{ nm}. \quad (7)$$

278 We note that the ratio of the **length to the film thickness is $d/z_0 \sim 2 \times 10^4$** . This high value of the
 279 ratio ensures that boundary effects are negligible, thereby also lending credibility to the statistical
 280 approach leading to Eq. 6.

281 The present value of the thermalization length $\langle z_0 \rangle$ turns out to be 3 to 5 times longer than in
 282 liquid helium at approximately the same temperature⁴⁴.

283 A lack of knowledge of the electron drift mobility and/or scattering cross section in the solid
 284 p-H₂ at the low temperature of our experiment does not allow us to gather some deeper pieces
 285 of information regarding the electron thermalization time that may be useful for the design of the
 286 experiment aimed at the neutralization of cations embedded in the solid.

287 Nevertheless, we conclude that thermalization of quasi-free electrons occurs well within the
 288 bulk. Upon thermalization, the electron thermal speed at $T = 2.8 \text{ K}$ is $v_{\text{th}} \approx 11 \times 10^3 \text{ m/s}$ to be
 289 compared to the average value of the sound speed $v_S \approx 2 \times 10^3 \text{ m/s}$ ^{54–57}. Thus, it is reasonable to
 290 assume that polarization of the medium via phonons is too slow to respond to the electron motion,
 291 which could otherwise hinder the Coulomb interaction of the electrons with the embedded BaF⁺
 292 cations.

293 **IV. CONCLUSIONS**

294 For the first time we have carried out measurements of the thermalization length of quasi-free
295 electrons in solid para-hydrogen at low temperature. The goal is to investigate their energetics and
296 dynamics in order to ascertain the possibility to neutralize cations embedded in a solid matrix of
297 this material.

298 The outcome of the present experiment has several facets. First of all, electrons of low energy
299 are injected into the conduction band of the solid by exploiting the photoelectric effect of a gold
300 cathode irradiated with a short pulse of a UV laser whose photon energy is just close to the dis-
301 sociation energy of H₂. By so doing, the production of very slow H⁻ anions is very unlikely. An
302 analysis of the time evolution of the waveforms we have recorded indicates that only fast negative
303 charges are present.

304 Secondly, we have been able to determine the thermalization length z_0 of quasi-free electrons.
305 It turns out that z_0 is quite large though well much shorter than the solid film thickness. This
306 is a useful piece of information regarding the possibility of neutralizing the cations that have to
307 be embedded in the solid p-H₂ matrix in order to carry out high-precision measurements of the
308 electric dipole moment of the electron.

309 However, the thermalization length of electrons in a matrix is an important quantity in sev-
310 eral more physics fields. It is well known that there might be a relationship between electron
311 thermalization and mobility^{58,59}. Moreover, among others, electron thermalization plays a role in
312 the physics of geminate ion recombination or of free ion yield which is important in the field of
313 radiation detectors^{60,61}, just to mention some applications.

314 In this first experiment we have investigated the electron thermalization at one single temper-
315 ature and density. We had to assume that there is a barrier to the injection of electrons into the
316 solid without actually measuring it because its determination requires a detailed knowledge of the
317 density dependence of the thermalization process. Moreover, it was impossible, in the present
318 experimental configuration, to measure the electron drift time. Therefore, we are planning for the
319 near future to extend our measurement campaign to fill in this gap.

320 **ACKNOWLEDGMENTS**

321 We wish to thank dr. D. Sali, Prof. A. C. Vutha, Prof. T. Momose, Prof. J. D. Weinstein,
322 and Prof. C. L. Caesar for helpful suggestions. A. F. Borghesani gratefully acknowledges useful
323 discussions with Prof. A. G. Khrapak, Prof. L. P. Mezhov-Deglin, and Prof. J.-P. Jay-Gerin.
324 We also express our thanks to E. Berto, F. Calaan, M. Tessaro, M. Rebeschini, and M. Zago
325 for technical support. G. Messineo acknowledges financial support from the European Union's
326 Horizon2020 research and innovation programme under the Marie Skłodowska-Curie grant no.
327 754496.

328 **AUTHOR DECLARATIONS**

329 **Conflict of Interest**

330 The authors have no conflicts to disclose.

331 **Authors contributions**

332 All authors equally contributed to the experiment.

333 **Data availability**

334 The data that support the findings of this study are available from the corresponding author
335 upon reasonable request.

336 **REFERENCES**

337 ¹A. C. Vutha, W. C. Campbell, Y. V. Gurevich, N. R. Hutzler, M. Parsons, D. Patterson, E. Petrik,
338 B. Spaun, J. M. Doyle, G. Gabrielse, and D. DeMille, *J. Phys. B: Atomic, Molecular and Optical*
339 *Physics* **43**, 074007 (2010).

340 ²P. Aggarwal, H. L. Bethlem, A. Borschevsky, M. Denis, K. Esajas, P. A. B. Haase, Y. Hao,
341 S. Hoekstra, K. Jungmann, T. B. Meijknecht, M. C. Mooij, R. G. E. Timmermans, W. Ubachs,
342 L. Willmann, A. Zapara, and the NL-eEDM collaboration, *Eur. Phys. J. D* **72**, 197 (2018).

- 343 ³V. Andreev, D. G. Ang, D. DeMille, J. M. Doyle, G. Gabrielse, J. Haefner, N. R. Hutzler,
344 Z. Lasner, C. Meisenhelder, B. R. O’Leary, C. D. Panda, A. D. West, E. P. West, X. Wu, and
345 ACME Collaboration, *Nature* **562**, 355 (2018).
- 346 ⁴P. Sandars, *Physics Letters* **14**, 194 (1965).
- 347 ⁵M. Guarise, C. Braggio, R. Calabrese, G. Carugno, A. Dainelli, A. Khanbekyan, E. Luppi,
348 E. Mariotti, M. Poggi, and L. Tomassetti, *Review of Scientific Instruments* **88**, 113303 (2017).
- 349 ⁶M. Guarise, C. Braggio, R. Calabrese, G. Carugno, A. Dainelli, A. Khanbekyan, E. Luppi,
350 E. Mariotti, and L. Tomassetti, *Nuclear Instruments and Methods in Physics Research Section*
351 *A: Accelerators, Spectrometers, Detectors and Associated Equipment* **936**, 244 (2019).
- 352 ⁷I. F. Silvera, *Rev. Mod. Phys.* **52**, 393 (1980).
- 353 ⁸A. Driessen and I. F. Silvera, *Phys. Rev. B* **35**, 6649 (1987).
- 354 ⁹H. Meyer, *Low Temp. Phys.* **24**, 381 (1998).
- 355 ¹⁰T. R. Prisk, R. T. Azuah, D. L. Abernathy, G. E. Granroth, T. E. Sherline, P. E. Sokol, J. Hu, and
356 M. Boninsegni, *Phys. Rev. B* **107**, 094511 (2023).
- 357 ¹¹A. A. Levchenko and L. P. Mezhov-Deglin, *Sov. Phys.-JETP* **71**, 196 (1990).
- 358 ¹²A. A. Levchenko, L. P. Mezhov-Deglin, and I. E. Shtinov, *JETP Lett.* **54**, 234 (1991).
- 359 ¹³P. G. Le Comber, J. B. Wilson, and R. J. Loveland, *Solid St. Comm.* **18**, 377 (1976).
- 360 ¹⁴A. A. Levchenko and L. P. Mezhov-Deglin, *JETP Lett.* **48**, 442 (1988).
- 361 ¹⁵A. A. Levchenko and L. P. Mezhov-Deglin, *J. Low. Temp. Phys.* **89**, 457 (1992).
- 362 ¹⁶A. A. Levchenko and L. P. Mezhov-Deglin, *JETP Lett.* **60**, 470 (1994).
- 363 ¹⁷P. B. Lerner and I. M. Sokolov, *J. Low Temp. Phys.* **95**, 683 (1994).
- 364 ¹⁸A. V. Berezhnov, A. G. Khrapak, E. Illenberger, and W. F. Schmidt, *High Temp.* **41**, 425 (2003).
- 365 ¹⁹A. F. Borghesani, *Ions and Electrons in Liquid Helium*, Int. Series of Monographs on Physics,
366 Vol. 137 (OUP, Oxford (UK), 2007).
- 367 ²⁰Y. Sakai, H. Böttcher, and W. F. Schmidt, *J. Jpn. Inst. Electr. Eng. A* **61**, 499 (1983).
- 368 ²¹A. G. Khrapak, W. F. Schmidt, and K. F. Volykhin, *Phys. Rev. E* **51**, 4804 (1995).
- 369 ²²R. J. Corruccini, “Surface tensions of normal and para hydrogen,” Technical Note 322 (Natl.
370 Bureau Standards, Boulder, 1965).
- 371 ²³M. H. Cohen and J. Jortner, *Phys. Rev.* **180**, 238 (1969).
- 372 ²⁴K. O. Keshishev, L. P. Mezhov-Deglin, and A. I. Shal’nikov, *Sov. Phys. JETP Lett.* **12**, 160
373 (1971).
- 374 ²⁵G. A. Sai-Halasz and A. J. Dahm, *Phys. Rev. Lett.* **28**, 1244 (1972).

- 375 ²⁶V. E. Dionne, R. A. Young, and C. T. Tomizuka, *Phys. Rev. A* **5**, 1403 (1972).
- 376 ²⁷V. B. Shikin, *Sov. Phys. Usp.* **20**, 226 (1977).
- 377 ²⁸A. F. Andreev and A. D. Savishchev, *Sov. Phys. JETP* **69**, 630 (1989).
- 378 ²⁹C. Ebner and C. C. Sung, *Phys. Rev. A* **5**, 2625 (1972).
- 379 ³⁰D. Zhou, M. Rall, J. P. Brison, and N. S. Sullivan, *Phys. Rev. B* **42**, 1929 (1990).
- 380 ³¹T. Kumada, H. Inagaki, T. Nagasawa, Y. Aratono, and T. Miyazaki, *Chem. Phys. Lett.* **251**, 219
381 (1996).
- 382 ³²T. Miyazaki, H. Fukuta, M. Hanabusa, T. Kumada, and J. Kumagai, *Chem. Phys. Lett.* **360**, 8
383 (2002).
- 384 ³³A. B. Trusov, L. P. Mezhov-Deglin, and A. A. Levchenko, *JETP Lett.* **63**, 376 (1996).
- 385 ³⁴B. A. Tom, S. Bhasker, Y. Miyamoto, T. Momose, and B. J. McCall, *Rev. Sci. Instrum.* **80**,
386 016108 (2009).
- 387 ³⁵K. Sundararajan, K. Sankaran, N. Ramanathan, and R. Gopi, *J. Mol. Struct.* **1117**, 181 (2016).
- 388 ³⁶A. Bhandari, A. P. Rollings, L. Ratto, and J. D. Weinstein, *Rev. Sci. Instrum.* **92**, 073202 (2021).
- 389 ³⁷M. E. Fajardo, “Physics and Chemistry at Low Temperatures,” (Pan Stanford Publishing, Singa-
390 pore, 2011) Chap. Matrix isolation spectroscopy in solid parahydrogen: A primer., pp. 167–202.
- 391 ³⁸M. E. Fajardo, *Appl. Spectrosc.* **73**, 1403 (2019).
- 392 ³⁹B. A. Younglove, *J. Chem. Phys.* **48**, 4181 (1968).
- 393 ⁴⁰J. H. Constable, C. F. Clark, and J. R. Gaines, *J. Low. Temp. Phys.* **21**, 599 (1975).
- 394 ⁴¹R. W. Crompton and A. G. Robertson, *Aust. J. Phys.* **24**, 543 (1971).
- 395 ⁴²A. G. Robertson, *Aust. J. Phys.* **24**, 445 (1971).
- 396 ⁴³J. W. Leachman, R. T. Jacobsen, S. G. Penoncello, and E. W. Lemmon, *Journal of Physical and*
397 *Chemical Reference Data* **38**, 721 (2009).
- 398 ⁴⁴D. G. Onn and M. Silver, *Phys. Rev* **183**, 295 (1969).
- 399 ⁴⁵D. G. Onn and M. Silver, *Phys. Rev A* **3**, 1773 (1971).
- 400 ⁴⁶P. Smejtek, M. Silver, K. S. Dy, and D. G. Onn, *J. Chem. Phys.* **59**, 1374 (1973).
- 401 ⁴⁷D. J. Griffiths, *Introduction to Electrodynamics* (Cambridge University Press, Cambridge, 2017).
- 402 ⁴⁸J.-S. Yoon, M.-Y. Song, J.-M. Han, S. H. Hwang, W.-S. Chang, B. Lee, and Y. Itikawa, *J. Phys.*
403 *Chem. Ref. Data* **37**, 913 (2008).
- 404 ⁴⁹L. Sanche, *Int. J. Radiat. Appl. Instrum. Part C. Radiat. Phys. Chem.* **32**, 269 (1988).
- 405 ⁵⁰L. A. Young and N. E. Bradbury, *Phys. Rev.* **43**, 34 (1933).
- 406 ⁵¹M. Silver, P. Kumbhare, P. Smejtek, and D. G. Onn, *J. Chem. Phys.* **52**, 5195 (1970).

Electron thermalization length in solid para-hydrogen at low-temperature

- 407 ⁵²J. R. Broomall, W. D. Johnson, and D. G. Onn, Phys. Rev. **B 14**, 2819 (1976).
- 408 ⁵³E. M. Gushchin, A. A. Kruglov, and I. M. Obodovskii, Sov. Phys. JETP **55**, 860 (1982).
- 409 ⁵⁴K. Kuroda, J. Z. Li, M. Suzuki, M. Katsuragawa, and K. Hakuta, J. Low Temp. Phys. **125**, 39
410 (2001).
- 411 ⁵⁵P. A. Bezuglyi, R. O. Plakhotin, and L. M. Tarasenko, Sov. Phys. Solid State **13**, 250 (1971).
- 412 ⁵⁶R. Wanner and H. Meyer, Phys. Lett. A **41**, 189 (1972).
- 413 ⁵⁷R. Wanner and H. Meyer, J. Low Temp. Phys. **11**, 715 (1973).
- 414 ⁵⁸H. Sano and A. Mozumder, J. Chem. Phys. **66**, 689 (1977).
- 415 ⁵⁹G. R. Freeman, “Kinetics of nonhomogeneous processes,” (Wiley, New York, 1987) Chap.
416 Ionization and charge separation in irradiated materials, pp. 19–87.
- 417 ⁶⁰E. Aprile, A. E. Bolotnikov, A. I. Bolozdynya, and T. Doke, *Noble Gas Detectors* (Wiley-VCH,
418 Weinheim (RFG), 2006).
- 419 ⁶¹C. Ferradini and J.-P. Jay-Gerin, *Excess Electrons in Dielectric Media* (CRC, Boca Raton (USA),
420 1991).