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Electron Photo-ejection from Bubble States in Liquid ^{4}He

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Abstract Within finite-range density-functional theory, we have addressed the photo-ejection of electrons hosted in ^{4}He bubbles as a function of pressure at zero temperature. It is shown that, besides the $1s \rightarrow 1p$ and $1s \rightarrow 2p$ transitions that show up in the whole pressure range up to solidification, there is another transition to a loosely bound $3p$ state that disappears at a pressure P above 1.7 bar. Realistic predictions for the electron photo-ejection cross section are made using a model that has been proven to reproduce the experimental infrared absorption transitions as a function of pressure.

Keywords Electron bubbles · Photo-ejection cross section · Liquid helium

1 Introduction

Electron bubbles (e-bubbles) produced by electrons injected into liquid helium are the subject of an extensive literature, see e.g. Refs. [1–3] and references therein. The change in the drift properties of excess electrons in liquid He submitted to electromagnetic radiation was studied long ago [4–6], constituting an experimental evidence of the existence of e-bubbles.

The absorption spectrum of e-bubbles has been experimentally determined with high accuracy [7, 8]. It is dominated by two discrete transitions, namely $1s \rightarrow 1p$ and $1s \rightarrow 2p$, the sum of whose calculated oscillator strengths is nearly unity, thus

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almost exhausting the Thomas-Reiche-Kuhn or energy weighted sum rule for the dipole operator [9, 10].

The theoretical description of e-bubbles was first based on the application of the simplest version of the bubble model characterized by an abrupt interface and a surface tension, see e.g. Refs. [11–13]. Key ingredients of this model are the surface tension of the helium free surface, only known along the saturation vapor curve, and the electron-He confining potential. In recent years, available accurate finite-range density functionals (DF) for liquid ^{4}He [14] and Hartree-type electron-He effective potentials [15, 16] have allowed for a detailed description of e-bubbles, in particular of their absorption spectrum [17, 18]. These and other DF calculations, such as those of Refs. [19–23] have established the suitability of DF theory to address inhomogeneous liquid helium, and have stressed the relevance of a correct description of the surface thickness of the e-bubble to achieve a quantitative description of the experimental data.

As mentioned, very little oscillator strength is left for transitions to bound np states with $n > 2$ and for those leading to electron photo-ejection. The cross section for electron photo-ejection was calculated long ago using the bubble model [13, 24]. In particular, the total oscillator strength for this process was determined to amount just a few parts in one thousand [13]. Thus, experiments aiming at studying electron photo-ejection from bubble states are challenging. The cross section for this effect was thought to have been measured at pressures from 10.5 to 249 psi [6] (we recall that 1 psi ~ 0.070 bar). However, in the analysis of these results the strength arising from the $1s \rightarrow 2p$ transition was mistakenly considered as part of the photo-ejection cross section, as discussed in Ref. [13].

In this work we present calculations of the photo-ejection cross section as a function of pressure at zero temperature, based on a model that has been able to reproduce the experimental infrared absorption transitions as a function of pressure. It uses a density functional approach to accurately describe the density profile of the e-bubble, and an accurate Hartree-type electron-He interaction. We also calculate the contribution of a discrete $1s \rightarrow 3p$ transition that appears at low pressures. The calculated oscillator strength is exhausted by the sum of the contributions from these discrete transitions plus the continuum photo-ejection contribution.

2 Method and Results

We have used the Orsay-Trento density functional [14] and the electron-He interaction derived by Cheng et al. [15, 16] This allows us to write the energy of the electron-helium system as a functional of the electron wavefunction $\Phi(\mathbf{r})$ and the 'effective wavefunction' $\Psi(\mathbf{r}) = \sqrt{\rho(\mathbf{r})}$, where $\rho(\mathbf{r})$ is the atomic density of ^{4}He. Varying the functional with respect to Ψ and Φ one obtains a system of two coupled equations, an Euler-Lagrange equation for the liquid and a Schrödinger equation for the electron, that have to be solved selfconsistently [25].

Figure 1 shows, at $P = 0$, the bubble density profile and the electron probability density for the $1s, 1p, 2p$ and $3p$ states, together with the confining potential well and single-electron energy levels. It is worth noticing the appearance of a bound

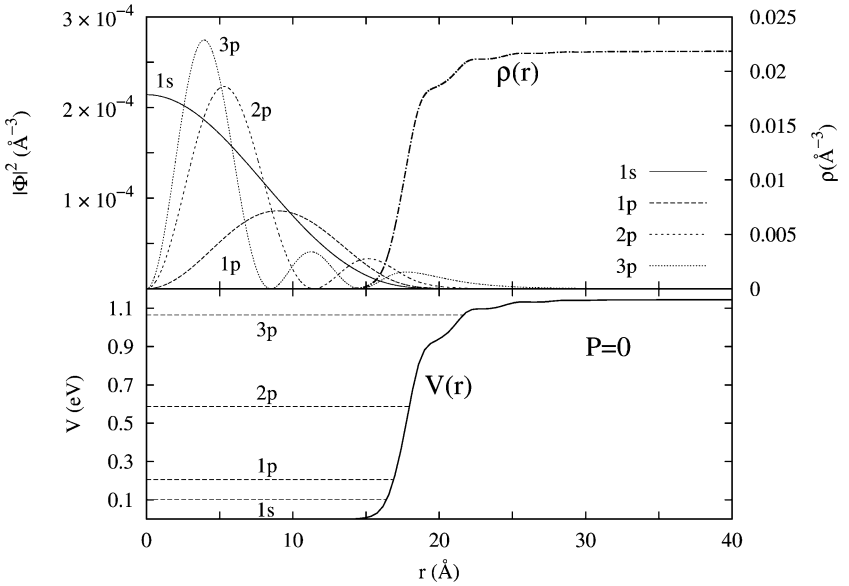

Fig. 1 *Top panel*: bubble atomic density profile (*right scale*) and electron probability density for the $1s, 1p, 2p$ and $3p$ states (*left scale*) at $P = 0$. *Bottom panel*: confining potential well and single-electron energies

$3p$ level that becomes unbound above $P = 1.7$ bar. The existence of this level was overlooked in Ref. [18]. The $3p$ state appears to be loosely bound, its energy being barely below the delocalization limit. Due to the approximated way used to describe the electron-helium interaction, the band-edge for ^{4}He appears to be slightly above 1.1 eV, see Fig. 1, whereas the experimental value is around 1.05 eV. This is a tiny energy difference which does not affect the main conclusions of this work. It could however have a sensible effect on the $3p$ state, which might not even be bound if a slightly less binding potential were used.

Photoabsorption cross sections have been obtained from first-order perturbation theory, assuming unpolarized photons with energy $W = \hbar\omega$ and wave vector $\mathbf{k}$ ($k = \omega/c$). The cross section for excitation from a state Φ_a of energy E_a to bound states Φ_b of energy E_b is given by

$$\frac{\mathrm{d}\sigma_{ba}^{\mathrm{exc}}}{\mathrm{d}W} = \frac{(2\pi)^2 e^2 c\hbar}{W} {\sum}' \left| \langle \Phi_b | \frac{1}{\mathrm{m_e} c} \exp(\mathrm{i}\,\mathbf{k}\cdot\mathbf{r}) \epsilon_\alpha \cdot \left[\mathbf{p} + \mathrm{i}\frac{\hbar}{2}\boldsymbol{\sigma} \times \mathbf{k} \right] | \Phi_a \rangle \right|^2 \times \delta(E_b - E_a - W) \tag{1}$$

where the primed summation indicates a sum over degenerate final states and averages over states of the initial level and over photon polarizations ϵ_α. The total cross section for absorption of photons with energy $W > -E_a$ (meaning electron ejection, as energies are here referred to the asymptotic value of the confining potential well) is

$$\sigma_{ba}^{\mathrm{ejec}} = \frac{(2\pi)^2 e^2 c\hbar}{W} \frac{k_b}{\pi E_b} {\sum}' \left| \langle \Phi_b^{(-)} | \frac{1}{\mathrm{m_e} c} \exp(\mathrm{i}\,\mathbf{k}\cdot\mathbf{r}) \epsilon_\alpha \cdot \left[\mathbf{p} + \mathrm{i}\frac{\hbar}{2}\boldsymbol{\sigma} \times \mathbf{k} \right] | \Phi_a \rangle \right|^2 \tag{2}$$

where the final state $\Phi_b^{(-)}$ is a distorted plane wave with incoming spherical distortion, and the primed summation includes an integral over the final direction of the emitted photoelectron; k_b is the wave number of the photoelectron. The constants in the above equation correspond to distorted plane waves normalized in wave-vector space.

To calculate these cross sections we adapted a Fortran code for atomic photoionization, which follows the theoretical formulation of Refs. [26, 27]. Although this program utilizes electron Dirac wavefunctions, in the case of e-bubbles it yields results that do not differ significantly from a pure non-relativistic calculation because of the smallness of the electron binding energy. Radial wavefunctions have been computed using the subroutine package RADIAL [28], which allows a strict control of truncation errors. A comparison with photoelectric cross sections for hydrogenic ions shows that the calculated cross sections are accurate to about 5 significant digits.

In the photon long-wavelength limit, the cross section reduces to the familiar dipole approximation expression

$$\sigma_{ba} = \frac{2\pi^2 e^2 \hbar}{\mathrm{m_e} c} \frac{\mathrm{d}f(W)}{\mathrm{d}W}, \tag{3}$$

where $\mathrm{d}f/\mathrm{d}W$ is the optical oscillator strength

$$\frac{\mathrm{d}f(W)}{\mathrm{d}W} = \frac{W 2\mathrm{m_e}}{\hbar^2} \sum_{E_b} \delta(E_b - E_a - W) \sum{}' \left| \langle \Phi_b | z | \Phi_a \rangle \right|^2 + \frac{W 2\mathrm{m_e}}{\hbar^2} \frac{k_b}{\pi E_b} \sum{}' \left| \langle \Phi_b^{(-)} | z | \Phi_a \rangle \right|^2 \tag{4}$$

which satisfies the sum rule

$$\int_0^\infty \frac{\mathrm{d}f(W)}{\mathrm{d}W} \mathrm{d}W = 1. \tag{5}$$

Table 1 shows the results we have obtained for the discrete $1s \rightarrow np$ transitions at selected pressures. For $n = 1$ and 2 they coincide with those obtained by Grau et al. [18]. Two new items are collected in the table, namely the results for the $1s \rightarrow 3p$ transition and the oscillator strength of the photo-ejection cross section up to 5 eV excitation energy. Notice that just a minute $2 - 3$ per thousand of the oscillator strength is concentrated in the photo-ejection cross section.

Figure 2 shows several photo-electron cross sections at different pressures. The cross section is shifted to higher energies as P increases, i.e., the radius of the e-bubble decreases. An approximate dependence of the cross section on the radius R of the e-bubble can be obtained from the pressure dependence of R shown in Fig. 5 of Ref. [18]. The calculated cross sections are qualitatively similar to those found by Wang [24] using the bubble model, although with significant differences likely due to the very different density profiles.

3 Summary and Outlook

Within finite-range density functional theory, we have carried out a study of the photo-ejection cross section of electron bubbles in liquid ^{4}He at zero temperature as a function of pressure.

Table 1 Properties of the discrete $1s \rightarrow np$ transitions at selected P values. The oscillator strength for photo-ejection below 5 eV energy is also indicated

P (bar)	Transition	Transition energy (eV)	Oscillator strength	Total cross section (eV cm^2)
0	$1s$–$1p$	0.104	0.9714	1.066×10^{-16}
	$1s$–$2p$	0.486	0.0250	2.747×10^{-18}
	$1s$–$3p$	0.963	0.0029	3.246×10^{-19}
	Photo-ejection		0.0007	
1	$1s$–$1p$	0.115	0.9718	1.067×10^{-16}
	$1s$–$2p$	0.533	0.0249	2.738×10^{-18}
	$1s$–$3p$	1.026	0.0020	2.266×10^{-19}
	Photo-ejection		0.0012	
1.6	$1s$–$1p$	0.121	0.9720	1.067×10^{-16}
	$1s$–$2p$	0.561	0.0248	2.731×10^{-18}
	$1s$–$3p$	1.053	0.0012	1.306×10^{-19}
	Photo-ejection		0.0019	
2.2	$1s$–$1p$	0.127	0.9723	1.067×10^{-16}
	$1s$–$2p$	0.587	0.0248	2.725×10^{-18}
	Photo-ejection		0.0027	
3	$1s$–$1p$	0.133	0.9725	1.068×10^{-16}
	$1s$–$2p$	0.615	0.0247	2.719×10^{-18}
	Photo-ejection		0.0028	
5	$1s$–$1p$	0.147	0.9729	1.068×10^{-16}
	$1s$–$2p$	0.676	0.0246	2.705×10^{-18}
	Photo-ejection		0.0025	
10	$1s$–$1p$	0.175	0.9735	1.069×10^{-16}
	$1s$–$2p$	0.793	0.0243	2.674×10^{-18}
	Photo-ejection		0.0021	
15	$1s$–$1p$	0.196	0.9739	1.069×10^{-16}
	$1s$–$2p$	0.883	0.0241	2.647×10^{-18}
	Photo-ejection		0.0019	
20	$1s$–$1p$	0.214	0.9742	1.070×10^{-16}
	$1s$–$2p$	0.958	0.0238	2.629×10^{-18}
	Photo-ejection		0.0019	

We believe that our method is reliable, as it has recently allowed us to reproduce the experimental $1s \rightarrow 1p$ and $1s \rightarrow 2p$ transition energies, and it overcomes the more relevant limitations of previous works, based either on too rough a description of the bubble density profile (sharp densities), the guessing of the surface tension of the helium free surface (only known along the saturation vapor line), too simple a

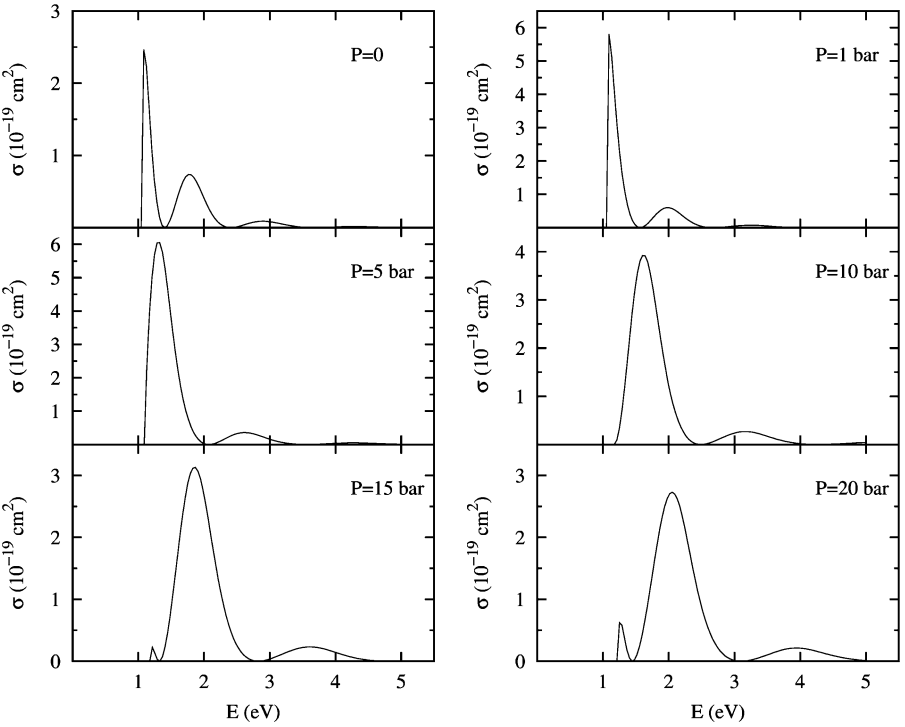

Fig. 2 Photo-ejection cross section as a function of energy at different pressures

model for the electron-He interaction, or on a combination of all these effects. In this sense, the results presented here might be a realistic guide to an accurate experimental determination of the electron photo-ejection cross section from e-bubbles, which despite the efforts made in the past is still an open problem due to the difficulties involved in its determination.

The present study can be naturally extended to ^{4}He at finite temperature, and to e-bubbles in liquid ^{3}He and in ^{3}He-^{4}He liquid mixtures at zero temperature, as accurate DF are available for these systems [29, 30]. Work along these lines is now in progress.

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References

1. M. Rosenblit, J. Jortner, J. Chem. Phys. **124**, 194505 (2006)
2. M. Rosenblit, J. Jortner, J. Chem. Phys. **124**, 194506 (2006)
3. H.J. Maris, J. Phys. Soc. Jpn. **77**, 1 (2008)
4. J.A. Northby, T.M. Sanders, Phys. Rev. Lett. **18**, 1184 (1967)
5. C. Zipfel, T.M. Sanders, in *Proceedings of the 11th Int. Conference on Low Temp. Phys*, ed. by J.F. Allen, D.M. Finlayson, D.M. McCall, St. Andrews, Scotland (1969), p. 296
6. C. Zipfel, PhD thesis, University of Michigan, unpublished (1969)

7. C.C. Grimes, G. Adams, Phys. Rev. B **41**, 6366 (1990)
8. C.C. Grimes, G. Adams, Phys. Rev. B **45**, 2305 (1992)
9. M. Weissbluth, *Atoms and Molecules* (Academic Press, New York, 1978)
10. E. Lipparini, *Modern Many-Particle Physics* (World Scientific, Singapore, 2008)
11. W.B. Fowler, D.L. Dexter, Phys. Rev. **176**, 337 (1968)
12. B. DuVall, V. Celli, Phys. Rev. **180**, 276 (1969)
13. T. Miyakawa, D.L. Dexter, Phys. Rev. A **1**, 513 (1970)
14. F. Dalfovo, A. Lastri, L. Pricaupenko, S. Stringari, J. Treiner, Phys. Rev. B **52**, 1193 (1995)
15. E. Cheng, M.W. Cole, M.H. Cohen, Phys. Rev. B **50**, 1136 (1994)
16. E. Cheng, M.W. Cole, M.H. Cohen, Erratum. J. Chem. Phys. **50**(16), 134 (1994)
17. J. Eloranta, V.A. Apkarian, J. Chem. Phys. **117**, 10139 (2002)
18. V. Grau, M. Barranco, R. Mayol, M. Pi, Phys. Rev. B **73**, 064502 (2006)
19. J. Classen, C.-K. Su, M. Mohazzab, H.J. Maris, Phys. Rev. B **57**, 3000 (1998)
20. M. Pi, M. Barranco, R. Mayol, V. Grau, J. Low Temp. Phys. **148**, 43 (2007)
21. L. Lehtovaara, J. Eloranta, J. Low Temp. Phys. **148**, 43 (2007)
22. F. Ancilotto, M. Barranco, M. Pi, Phys. Rev. B **82**, 014517 (2010)
23. D. Mateo, D. Jin, M. Barranco, M. Pi, J. Chem. Phys. **134**, 044507 (2011)
24. S.-y. Wang PhD thesis, University of Michigan, unpubished (1967)
25. M. Pi, R. Mayol, A. Hernando, M. Barranco, F. Ancilotto, J. Chem. Phys. **126**, 244502 (2007)
26. R.H. Pratt, A. Ron, H.K. Tseng, Erratum. Rev. Mod. Phys. **45**, 663 (1973). Erratum, Ibid. 45 (1973)
27. J.H. Scofield, Technical report UCRL-51326, Lawrence Livermore Laboratory, Livermore, California (1973)
28. F. Salvat, J.M. Fernández-Varea, W. Williamson, Comput. Phys. Commun. **90**, 151 (1995)
29. F. Ancilotto, F. Faccin, F. Toigo, Phys. Rev. B **62**, 17035 (2000)
30. M. Barranco, M. Pi, S.M. Gatica, E.S. Hernández, J. Navarro, Phys. Rev. B **56**, 8997 (1997)