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Physica B 374–375 (2006) 355–358
PHYSICA B
www.elsevier.com/locate/physb

The effect of adsorbed noble gas atoms on muonium formation at the silica surface

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Abstract

Muonium formation on a silica powder surface has been studied while films of noble gases up to a monolayer thickness covered the surface of the powder grains. The fraction of muons stopped in the powder that formed muonium was found to decrease as the surface coverage increased for all the gases investigated. This has enabled muonium formation in the bulk, and at the surface of the grains, to be distinguished and provided the first direct evidence for the formation of muonium atoms at the surface of an insulator.

PACS: 36.10; 72.80.5k; 76.75.+i

Keywords: Muon-spin rotation; Muonium formation; Silica; Noble gases

1. Introduction

When muons are implanted into fine-grained silica powder large amounts of muonium atoms are formed with a good yield and emitted into the vacuum between the grains [1–5]. Recent developments of low-energy (eV–keV) muon beams for near-surface implantation studies [6–9] has reawakened interest in the surface interactions of both muons and muonium. In this paper, we study the effect of adsorbed noble gas atoms on the muonium formation probability in silica powder. This work improves upon and extends that was reported for helium on silica [5] and has afforded a more detailed insight into the mechanisms for muonium formation.

2. Experimental results

The experiments were performed on the EMU beamline at the ISIS pulsed muon facility [10]. The target was formed using Cab-O-Sil EH5® fumed silica powder (70 Å grain diameter) [11], which was mechanically pressed into a pellet of density around 0.2 g cm^{-3}. The pellet was baked in a vacuum oven at a temperature of 120 °C for several hours (typically overnight) before being loaded into a special sample cell in a glove box. The cell was connected to a gas line and gas-handling system and was inserted into a liquid helium cryostat. Noble gases He, Ne, Ar, Kr and Xe were introduced into the cell while the sample was at a temperature of 6, 20, 63, 95 and 154 K, respectively. The coverage, θ in monolayers, was determined by fitting the measured adsorbed volume as function of the equilibrium pressure at a constant temperature to the well known BET model [12,13]. The μSR experiments were performed at 6 K. The relaxations rate and the Mu asymmetry were subsequently extracted from the μSR spectra using standard fitting functions. The muonium asymmetry parameter as a function of the surface coverage is plotted in Fig. 1.

3. Discussion

For all the five noble gases, the asymmetry parameter drops rapidly as the surface gets covered and reaches a plateau-like region at $\theta = 0.4-0.5$. The faster than linear

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doi:10.1016/j.physb.2005.11.094

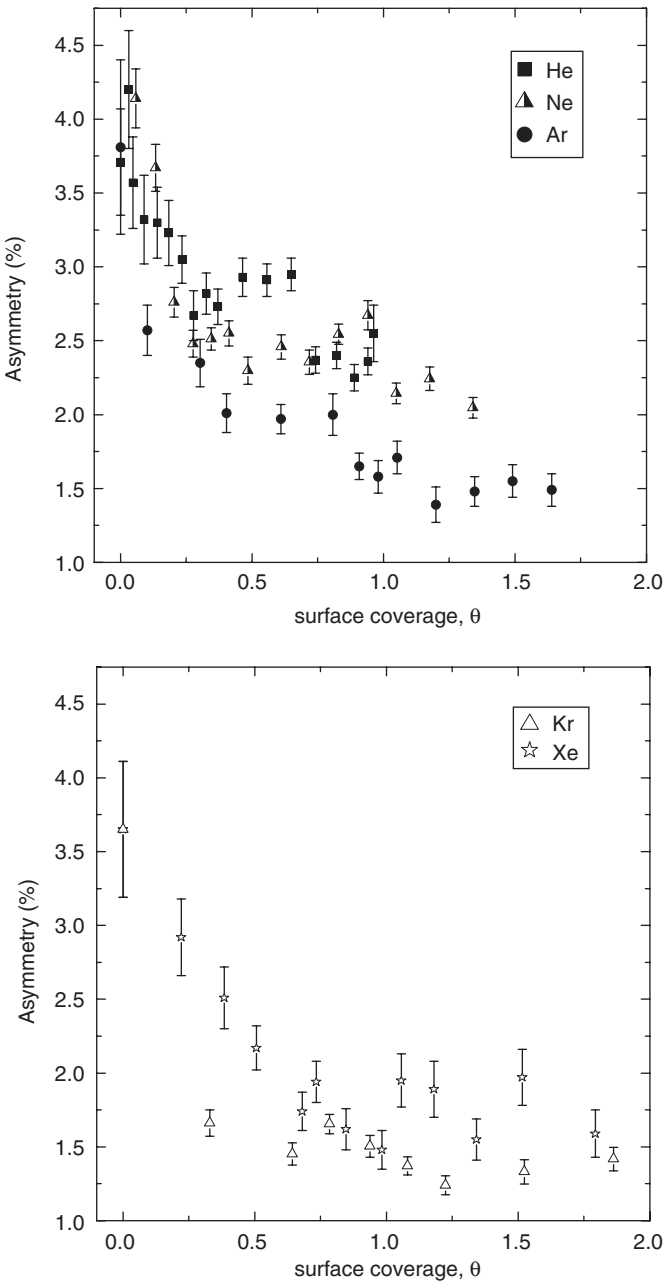

Fig. 1. Muonium asymmetry amplitude versus powder surface coverage for the noble gases.

drop with coverage seems to indicate that the gas is adsorbed preferentially at sites which are also active in promoting muonium formation. A second drops occurs when the coverage reaches a monolayer for all gases except xenon.

Previously, it was postulated from μSR experiments of helium on silica [5] that the drop in the asymmetry could either be caused by helium atoms inhibiting muonium formation at the surface or that there is charge exchange between the muonium and a positively charged helium ion. This ion could be formed in the trail of the stopping muons. The xenon ionization energy of about 12 eV however is lower than that of the muonium atom of about 13.6 eV. Thus charge transfer from muonium to ionized xenon is ruled out. It would also be very small in the case of krypton where the ionization energy is about 14 eV. Because the electron affinities of noble gases are negative Mu^- cannot be formed. Therefore the only option is that muonium formation can take place at the silica surface as well as in the bulk of the material. This result is similar to that found from a detailed study of the origins of positronium emitted into vacuum from a silica single crystal [14].

The muonium yields decrease with increasing coverage so we can conclude that part of the muonium is formed on the surface of the silica grains. The remaining asymmetry at $\theta = 1$ is attributed to muonium formation in the "bulk" of the powder grains [8,15]. The value of the asymmetry parameter decreases with increasing mass of the gas atom but is constant at around 1.5% for argon, krypton and xenon. From this we can infer that the lighter atoms still play a role in the muonium formation at the surface, likely involving their removal from the surface during the formation process. Most probably, the asymmetry at high coverages of argon, krypton and xenon reflects the true muonium formation in the bulk.

Electrons trapped at the surface, originating from the slowing down processes of the energetic muon, probably play a part in the formation of surface muonium. The number of surface sites for electrons will decrease with increasing coverage and consequently so will the surface yield of muonium. Bulk silica experiments have shown [15] that the stopped muon and the last track electrons are 10–15 nm apart. The typical length of the Cab–O–Sil EH5® grain aggregates is a round 200–300 nm [11]. Therefore the aggregates can be a source of localized electrons for muonium formation.

In Fig. 2 we show the measured spin-polarisation relaxation rates versus coverage for the five gases. The rate decreases with increasing coverage until a constant value below $0.5\,\mu s^{-1}$ is reached. For neon, argon and krypton the constant value is reached before $\theta = 1$, while for helium the change is slower. Thus, the adsorbed noble gas atoms reduce dramatically the depolarising effects of the bare powder surfaces. Naturally occurring xenon isotopes exhibit nuclear moments, which could explain its different behaviour.

Earlier measurements on muonium spin relaxation in Cab–O–Sil EH5® [3,5] attributed the relaxation to three mechanisms. Firstly, when the muonium atom diffuses past surface hydroxyl protons transitions are induced between the coupled muon-electron spin states [16]. Secondly, a different set of transitions within the muon-electron spin states is induced as the muonium atom visits different sites [17]. And thirdly there can be spin exchange with surface-trapped electrons.

From our data, we conclude that muonium formed both in the bulk and at the surface spends most of its life at, or near, the silica surface. This is in accordance with the results of low temperature studies of Harshman and

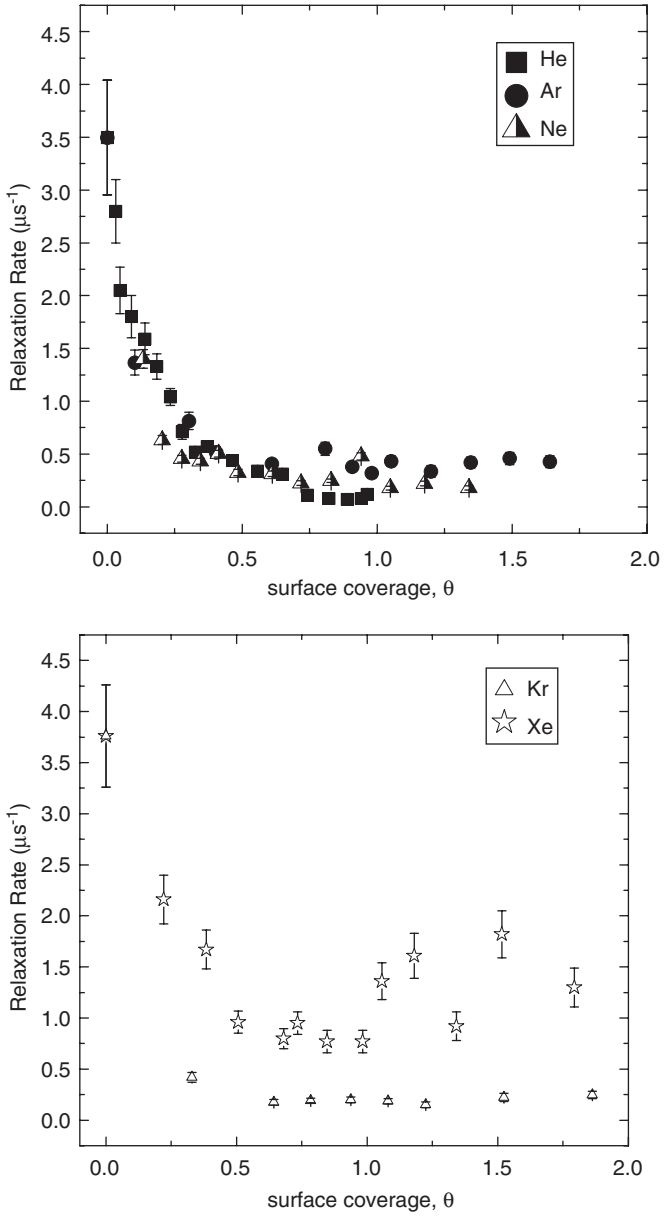

Fig. 2. Muonium spin-polarisation relaxation rates versus powder surface coverage for the noble gases.

co-workers [3,5]. Further support for surface-bound muonium was forthcoming from separate longitudinal field repolarisation experiments [18].

Fig. 3 shows that a proportion of the polarisation is regained in the presence of small longitudinal magnetic fields [19]. This seems to imply that there exists an anisotropic muonium state, as expected for a bound state at the surface. This was not observed at 300 K, corroborating previous studies [2–5]. Furthermore, the raised relaxation rate in the adsorbed xenon atoms is close to the measured rate in liquid xenon [20], implying that the muonium spends most of the time close to the xenon nuclei and, consequently, at or close to the silica surface.

The observation of surface-formed muonium is new. This has not been observed in the case of Al_2O_3

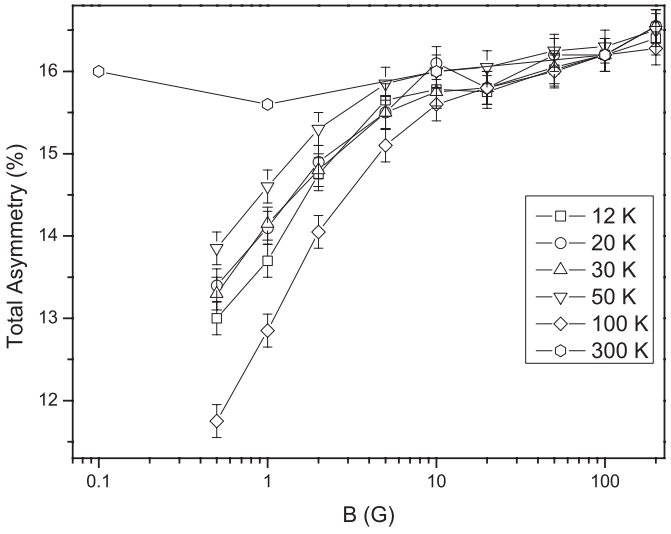

Fig. 3. Longitudinal repolarisation curves for the bare silica powder at various temperatures between 12 and 100 K, and at 300 K.

powders [2]. Interestingly, recent results have shown that positronium is formed at the surface and in the bulk of SiO_2, but not at the surface of KI and MgO [14]. If the muonium is formed in the bulk and is thermalised before emission, then its emission kinetic energy, T_B, can be written [14] as

$$T_B = -\phi_\mu - \phi_- - E_B + E_G + 13.6\,\text{eV}, \quad (1)$$

where ϕ_μ and ϕ_- are the muon and electron work functions, E_B is the binding energy of bulk muonium (13.6 eV is the vacuum value) and E_G is the silica band gap. On the other hand, if muonium is formed at the surface by capture of a valence electron by a thermalised muon the maximum emission energy will be

$$T^v_{\text{Surface}} = -\phi_\mu - \phi_- + 13.6\,\text{eV}. \quad (2)$$

From the known band gap and electron work functions for silica [14] and from the fact that $\phi_\mu > 0$ we can constrain (1) and (2). Using 11 and 10 eV for ϕ_- and E_G and since $T_B, T^v_{\text{Surface}} > 0$ we find, from (1), $(\phi_\mu + E_B) < 12.6$ eV and from (2), $\phi_\mu < 2.6$ eV. These (mild) conditions do not appear to be contradictory. Presumably muonium formed by either the bulk or surface processes will be ejected momentarily into the extragranular spaces.

Acknowledgements

PAD wishes to thank the EU, University College London (Peter Roberts Studentship) and the University of Wales Swansea for financial support. We thank ISIS and Prof. T. Hyodo for helpful communication.

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