Original Article

Bulletin of Pure and Applied Sciences.
Vol.37 D (Physics), No.2, 2018: P.163-166
Print version ISSN 0970 6569
Online version ISSN 2320 3218
DOI: 10.5958/2320-3218.2018.00022.2

Photoionization Spectroscopy of Lithium, Sodium and Potassium Nanoparticles in a Beam for the Metallic Work Function

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Received on 27.06.2018
Accepted on 15.11.2018

Abstract

We have used a modified particle source with a temperature controlled nozzle to measure the temperature dependence of the work function for Lithium, Sodium and Potassium. The temperature dependence of the work function arises from thermal expansion of the metal which resulted in a decrease of the electron density and a concomitant change of the Fermi energy. We have shown that photoionisation spectroscopy of Lithium, Sodium and Potassium nanoparticles in a beam provide high accuracy data for the metallic work function and its temperature dependence. The influence of temperature on the photoionization spectra of smaller clusters where thermal expansion has been predicted to have a stronger effect than in bulk and of particles composed of materials which exhibit measurable changes in the work function and photoyield at the melting point.

Keywords: Work function, electron density, thermal expansion, Fermi energy, nanoparticles, cluster, photoionization.

1. Introduction

KieJna and Pogosov [1] used semiempirical theories for incorporating thermal expansion effects have made specific predictions about the magnitude of the effect in various metals. Accurate measurements of work functions as function of temperature are therefore quite instructive. For most metals the Fermi temperature greatly exceeds the ambient temperatures. Michalelson [2] and Cadona and Ley [3] showed that it is not trivial to resolve such shift, because both the experimental precision and ab initio capability are typically more coarse than the effect. The temperature shift of the metal work function is an interesting subject because it concerns the interplay between structural e.g. phonon, excitation, thermal expansion, melting, and electronic degrees of freedom, the behavior of surface potentials. Gohlich et al. [4] and Wong et al [5] demonstrated that accurate measurements of metal work functions can be obtained by studying free nano particles rather than bulk surfaces. Kittel et al. [6] and Zaugwell [7] showed that the photoelectric effect of metals do not play much attention to possible variation of the surface work function with temperature. Whitefield et al. [8] presented the metal work function of alkalimetals. Burtscher and Siegmann [9] studied the temperature effect work function. Kasperovich et al. [10] showed that time of flight spectra indicated a distribution with average radius and a full width at half maximum of ~2nm. This average size is consistent with an earlier independent measurement based on electron attachment cross section. Saunders et al. [11] and Heer et al. [12] presented that once the clusters leave the heating tube, they undergo a 2 –m long free flight before being ionized by monochromatic light and subsequently detected by a Daly dynode photo multiplier ion counting arrangement. Durakiewicz et al. [13] provided a good fit for most metallic elements and evidently identified the main scaling variables as far as the temperature dependence of the work function was concerned.

2. Method

Accurate measurements of metal work functions can be obtained by studying free nanoparticles rather than bulk surfaces. In our setup a beam of nanoscale clusters several nm in diameter was generated in metal vapor condensation source, the particles are ionized by near-UV light of variable frequency and the resulting yield of positive ions was measured. This method takes advantage of the fact that the particle flight time in a molecular beam is very short in milliseconds and therefore surface contamination can be reduced to a minimum without a need to resort to ultrahigh vacuum surface preparation techniques. At the same time the use of ion-counting detection ensures high sensitivity. Large clusters of lithium, sodium and potassium are produced by evaporating metal from a crucible and quenching the vapor in a flow of cold helium gas. Nano-clusters condense in the aggregation zone, exit the source through a long nozzle length 25 nm, inner diameter 2 to 3.5 nm and for a collimated beam in free flight towards the defector. In order to control their temperature, a cylindrical thermalization tube was added to the setup. The tube which is 17 cm long with an inner diameter of 11 nm attached directly behind the nozzle. The temperature of the tube was controlled by electrical heater coils and monitored by three thermocouples ensuring uniform temperature distribution along the tube. It was maintained at any temperature between 300K and 500K with less than 2K temperature difference between the ends. The geometry of the tube was chosen to obtain thermal equilibrium between the tube and the clusters. Wiley-McLaren time of flight mass spectrometer was used for an overall characterization of the nanocluster population. It also served to gurard against the possibility of strong changes in the particle size distribution in response to varying source conditions. The time of flight spectra indicated a distribution with an average radius of 3 to 5 nm and full width at half maximum of ~2 nm.

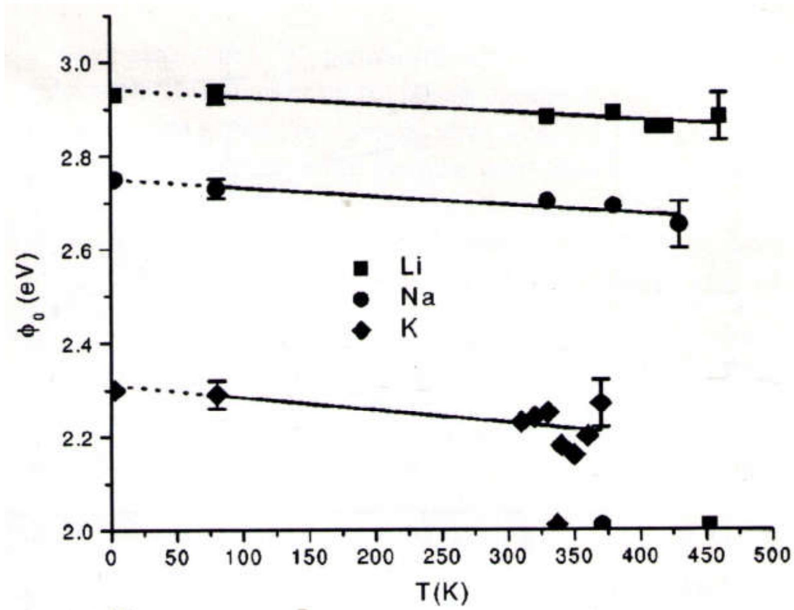
3. Results and Discussion

Graph (1) shows the measured temperature dependent work function for free nano particles of Lithium, Sodium and Potassium. The lines are linear least squares fit. The work functions extrapolate precisely to polycrystalline bulk surface work functions at zero temperature. This is the appropriate limit for our case. The randomly oriented particles present multiple nano scale faces to the light beam. In the case of polycrystalline bulk surfaces with patches the data can be described in terms of single effective work functions. The weak temperature induced work functions shifts were resolved quite reliably by the nano particle photo yield data. The Graph (1) also indicates that the position of the bulk melting point for each metal. No drastic changes were observed in either the work function or the shape of the photo yield curve upon crossing the bulk melting temperature. This is in agreement with previous work. Graph (2) shows a Fowler plot for some of our data on lithium, sodium and potassium nano particles. By varying ϕ_0 to optimize the fit we obtain the temperature dependent ionization potentials and the estimated accuracy of the fitted value. The equation for the photoelectron yield is given as

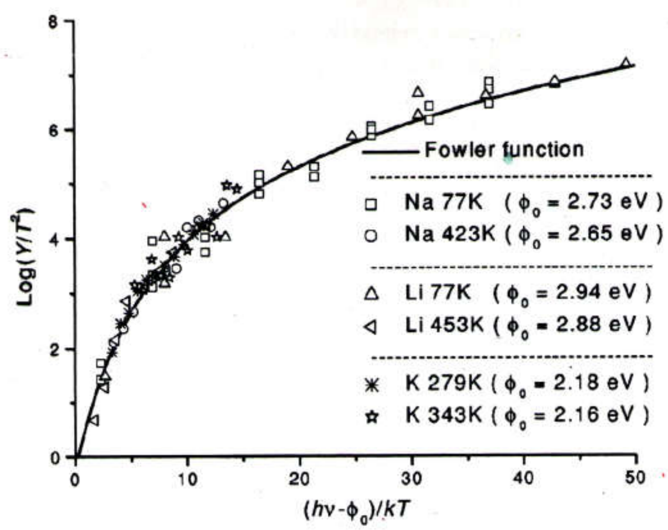
$$In\left(\frac{Y}{T^2}\right) = B + In\,f\left(\frac{h\nu - \phi_0}{k_B T}\right),$$

Where *f* is a known function

A plot of $In\left(\frac{Y}{T^2}\right)$ versus $\left(\frac{h\nu - \phi_0}{k_B T}\right)$ is known as Fowler plot and by a fit of the data to the universal curve we extracted the work function. It was found that the shape of the photoyield curve in the threshold region was not measurably altered.



Graph: 1: Work functions as a function of temperature. The solid straight lines are least-squares fits. The dotted lines are extrapolations to the polycrystalline work functions for bulk surfaces at zero temperature. The marks on the *x* axis indicate the bulk melting point for each metal.



Graph 2: Fowler plots for several selected yield curves. By varying ϕ_0 to align the data with the universal Fowler function *f*, the work function can be determined for various temperatures.

4. Conclusion

We studied the photoionization of lithium, sodium and potassium nano particles in a beam for the metallic work function and measured temperature dependent work functions for free nano particles of alkali metals. It was found that the temperature dependence of the work function arised primarily from thermal expansion of the metal which resulted in a increase of the electron density a concomitant change of the Fermi energy. It was found that the shape of the photo yield curve in the threshold region was not measurably altered. All the large particles in the distribution have the same threshold behavior; while the contribution of the smaller ones, shifted away to higher frequencies and is suppressed by their small cross sections. The weak temperature induced work functions shifts were resolved quite reliably by the nano particle photo yield data. The study of free nano particles in beams offers a very accurate complement to traditional surface spectroscopy. The obtained results were found in good agreement with previously observed and calculated values.

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