London Penetration Depth of Zero-Dimensional Pb Nanoparticles

P. J. Huang, M. K. Chung, C. C. Yang, and W.-H. Li*

Department of Physics, National Central University, Chung-Li, 32054 Taiwan

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The London penetration depths of zero-dimensional Pb nanoparticles were studied by measuring the ac magnetic susceptibility with a weak probing magnetic field. Both the diamagnetic screening effect that results in negative values for $\chi'$ and the dissipation peaks in $\chi''$ that signify the loss of the probing field to the screening current are clearly seen. The London penetration depth becomes shorter for smaller nanoparticles. A plot of the variation of the London penetration depth with particle size shows that the surface effect is less significant than the quantum size effect in reducing the penetration depth, and the quantum size effect becomes significant for particles smaller than 6 nm.

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It is known that an applied magnetic field would trigger a superconductor to setup a screening current on its surface to cancel the magnetic flux inside the superconductor and create an essentially flux free interior. However, magnetic flux would be present within a thin layer on the surface that supports the screening current. In the pure superconducting state, the magnetic field in the superconductor can generally be taken as being exponentially damped when it goes from the surface into the interior [1]. The London penetration depth $\lambda_L$ is the characteristic length that measures the thickness of the thin layer, on the surface of the superconductor, that supports the shielding current, in terms of how deep a magnetic field can penetrate into the superconductor. Typically, $\lambda_L$ takes a value of several tens of nms in a pure superconductor. In a zero-dimensional superconductor, where all of the size dimension are smaller than or comparable to the characteristic penetration depth and coherence length, dramatic changes in $\lambda_L$ can be expected [2, 3]. Here, we report on the results of studies made on the effects of particle diameter on $\lambda_L$.

Seven sets of Pb nanoparticle powders were fabricated by employing the standard gas-condensation method. Details of the fabrication processes can be found elsewhere [4]. The resultant nanoparticle powders, except for one, were no longer dull gray but dark black. They were in powder form, and were assemblies of macroscopic amounts of individual Pb nanoparticles. The samples were sensitive to being exposed to the air and were kept in a vacuum at all times to avoid surface oxidization. X-ray diffraction and atomic force microscopy (AFM) were used to determine the mean particle diameters and size distributions of the powders. All the diffraction peaks can be associated with an fcc Pb structure. No oxidation phases were found to within the 4 % x-ray resolution limitation. It appears that the diffraction peaks are much broader than those of the instrument resolution, reflecting the finite-size effect. Size analysis, based on the AFM images, shows that the size distribution can be described by using a Gaussian function, with a half-width typically of 1.5 nm.
FIG. 1: Plots of $\chi'(T)$ measured at various $H_a$ for the 6 nm particles. $\chi'(T)$ can be described by using the London penetration equation for small particles (solid lines).

FIG. 2: Plots of $\chi''(T)$ measured at various $H_a$ for the 6 nm particles. Dissipation peaks that signify the losses of the probing field to the screening currents are clearly seen.

for all the powders. The mean particle diameters determined for the seven sets of sample are 4.5, 6.0, 7.5, 9.0, 13.0, 16.0, and 86 nm. Note that bulk Pb has a superconducting transition temperature of $T_C = 7.2$ K, a London penetration depth at zero temperature of $\lambda_L(0) = 39$ nm, and a coherence length at zero temperature of $\xi(0) = 87$ nm [5]. The diameters for all powders fabricated are much smaller than the $\lambda_L$ and $\xi$ of bulk Pb. They may be classified as zero-dimensional superconductors.

London penetration depths of the nanoparticles were obtained by measuring the ac
FIG. 3: Plot of the variations of $\lambda_L$ with $H_a$ for various nanoparticles, showing that the rate of increase of $\lambda_L$ with $H_a$ is largely reduced for particles smaller than 6 nm.

magnetic susceptibility, with and without the presence of an applied dc magnetic field, $H_a$. To reduce the effects that may result from the probing field, the susceptibility measurements were performed by employing a weak probing ac field, with an rms strength of 1 Oe and a frequency of 100 Hz. It is known that the critical magnetic field for nanoparticles can be two orders-of-magnitude higher than that of bulk material [4]; the effects resulting from the probing field on the intrinsic superconducting properties are then insignificant. Figs. 1 and 2 show, respectively, the temperature dependencies of the in-phase component, $\chi'(T)$, and the out-of-phase component, $\chi''(T)$, of the magnetic susceptibility of the representative 6 nm powder, measured at various $H_a$. Diamagnetic screening effects that result in negative values for $\chi'$ at low temperatures are clearly seen in Fig. 1. Correspondingly, the dissipative losses of the probing field to the shielding currents, mainly within the individual nanoparticle, that result in a dissipation peak in $\chi''(T)$ are clearly revealed in Fig. 2. A significant drop in resistivity (not shown) was also observed at the corresponding temperature. These observations are characteristic of the occurrence of superconductivity. It appears that the transition occurs over a broad temperature range, as expected for zero-dimensional superconductors. The superconducting characteristic lengths of zero-dimensional nanoparticles are certainly limited by the particle size, that reduces the energy associated with the spatial fluctuations of the superconducting order parameter, and a broad critical transition, due to thermal fluctuations, may then be anticipated [6]. Although the diamagnetic screening effect is reduced when an $H_a$ is present, it is, however, still clearly evident at $H_a = 5.5$ T, which is already a factor of 70 times higher than the bulk critical magnetic field of $H_C=0.08$ T.

The diamagnetic susceptibility for small particles, where the effects originating from
FIG. 4: Plot of the dependence of $\lambda_L$ on particle diameter, showing that the quantum size effects become significant for particles smaller than 6 nm, below which $\lambda_L$ decreases abruptly.

the level quantization are not yet significant, can be obtained by applying the London equation to a small particle. For spherical particles of diameter $d$ with a mass density $\rho$, the diamagnetic susceptibility can be written as [7]

$$\chi'(T) = -\frac{3}{2\rho} \left\{ 1 - \frac{6\lambda_L(T)}{d} \coth \left( \frac{d}{2\lambda_L(T)} \right) + \frac{12\lambda_L^2(T)}{d^2} \right\},$$

assuming a London field penetration and that no flux vortices may appear further inside the interior of the particle. In the mean field regime, $\lambda_L$ varies with temperature according to

$$\lambda_L(T) = \lambda_L(0) \left[ 1 - \left( \frac{T}{T_C} \right)^4 \right]^{-1/2}.$$

All of the observed $\chi'(T)$ shown in Fig. 1 may be described (solid curves) by using the above expressions, signaling that no flux vortices appeared inside the particle even at $H_a = 4$ T, as was expected for type-I superconductor Pb. However, $\lambda_L$ becomes significantly shorter than that of bulk Pb. The $H_a$ dependence of the fitted $\lambda_L(0)$ for various nanoparticles are plotted in Fig. 3, revealing that $\lambda_L(0)$ reduces to 12.2(2) nm at $H_a=0$, and it increases to 31.2(5) nm at $H_a = 4$ T for the 6 nm particles. It is known that $H_a$ reduces the density of superconducting electrons; more volume, hence a longer penetration depth, is then needed in supporting the screening current when an $H_a$ is present. Variations of $\lambda_L(0)$ with $H_a$ for a fixed particle size may be anticipated. The observation that $\lambda_L(0)$ increases with an increasing $H_a$ shows that more energy is needed to shield a stronger $H_a$; that the rate of
increase of $\lambda_L$ with $H_a$ is largely reduced for particles smaller than 6 nm indicates that the quantum size effect has become significant.

The variation of $\lambda_L(0)$ with the mean particle diameter, $d$, is displayed in Fig. 4. It reveals an abrupt drop of $\lambda_L(0)$ at $d = 6$ nm, showing again that quantum size effects have become significant. It is known that quantum size effects are related to the appearance of the discrete nature of the electron levels near the Fermi energy. The sharp crossover from the bulk to quantum-confined regimes simply reflects the fact that the level separations increase rapidly as particles become smaller. This is understandable, since the average level separation is known to be inversely proportional to $d^3$ [8]. Furthermore, it is known that surface effects can become important for particles smaller than 50 nm. No significant reduction in $\lambda_L$ was observed for particles bigger than 10 nm, indicating that the surface effect is less significant than the quantum size effect in reducing the penetration depth.

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References

∗ Electronic address: whli@phy.ncu.edu.tw