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Optical Detection of the Superconducting Proximity Effect: Raman Scattering on Nb/InAs

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Raman scattering on InAs coated with 60 to 100Å thick Nb films (T_c = 2.5 to 5K) is studied. The Nb films, sputtered onto *in-situ* ion-etched n^+-InAs ($1.2\times10^{19}cm^{-3}$) are flat to ~5Å. The low-frequency coupled plasmon-phonon Raman mode (L_-) associated with bulk InAs is centered at 221cm^{-1}. The forbidden LO phonon at 237cm^{-1}, associated with the surface charge accumulation region (CAR), is observed because the thickness of the surface CAR is less than the Thomas-Fermi screening length. As the temperature is reduced below T_c the magnitude of the LO phonon mode intensity, relative to that of the L_- mode, increases by more than 40%

1. INTRODUCTION

Superconducting proximity effect studies in systems in which the normal metal is replaced by a semiconductor [1] are intriguing because the electronic properties (i.e., Fermi velocity, effective mass and carrier concentration) of the semiconductor can be varied with doping. Furthermore, the strength of the Schottky barrier which forms at the semiconductor interface can be engineered. For (100)-InAs this barrier is replaced with a charge accumulation region (CAR) so an ohmic contact forms at this interface.[2]

Raman spectroscopy is a powerful probe of near-surface electronic structure of semiconductors.[3,4] We have for the first time used Raman scattering to study the effect of superconducting and normal-state Nb films on the near-surface electronic properties of n^+-InAs. Much of this work is reported elsewhere.[5,6]

In earlier work, the crossover from quasiparticle tunneling to Andreev reflection was studied as a function of Schottky barrier thickness in Nb/InGaAs bilayers.[7] For high-transmittance junctions an excess conductance detected at low bias was attributed to the existence of a new current-carrying channel, Cooper-pair tunneling. This excess conductance was later taken as confirmation of the theory of "reflectionless tunneling", a mesoscopic effect which relies on a phase-coherent Andreev reflection producing single-quasiparticle phase coherence.[8] In order to differentiate between these models, which deviate in the high-transmittance regime, we have studied Nb/InAs, since n-(100)-InAs forms a charge accumulation region (CAR) at the interface, not a Schottky barrier.

Separate Raman modes, associated with the bulk and CAR, are observed. The coupled phonon-plasmon mode L_- is primarily associated with the bulk region, and the unscreened LO phonon mode is associated with the CAR. The latter is allowed because the magnitude of the wave vector of the LO phonon in the CAR is larger than the Thomas-Fermi wave vector.[9,10]

2. EXPERIMENTAL DETAILS

Thin Nb films are grown on single-crystal substrates of n^+-InAs ($n=1.2\times10^{19}cm^{-3}$) and nominally undoped InAs ($n=1.8\times10^{16}cm^{-3}$) by dc-magnetron sputter deposition after a gentle, *in-situ* Ar^+ etch of the substrate surface. X-ray reflectivity reveals the 100Å thick Nb films exhibit a surface roughness of less than 5Å and are featureless to the 30Å resolution of SEM.

For Nb films thinner than 2000Å, gradual reduction in T_c below the bulk 9.2K value is observed with decreased film thickness. Below 400Å the T_c drops dramatically with decreasing film thickness, and for 100 and 60Å films, T_c's are 5 and 2.5K, respectively.

Raman spectra are collected in a near-backscattering geometry using an Ar^+ laser focused on the sample with a cylindrical lens. A triple grating spectrometer equipped with a CCD camera is used to analyze the scattered radiation. The analysis is performed in the $x(y,z)\bar{x}$ configuration where the L_- mode is allowed [11], and in the $x(y,y)\bar{x}$ configuration where only the LO phonon is allowed. At temperatures near and below T_c, the Nb/InAs structures are immersed in superfluid helium while the temperature is monitored using a Pt-resistance thermometer buried in the copper block holding the sample.

3. RESULTS

In order to determine the thickness of the CAR and the effects of the Ar^+ etching and Nb deposition on the InAs, Raman spectra were taken at room temperature on InAs substrates before and after etching, and on Nb-coated InAs. Four excitation wavelengths were used. Two (488, 514.5nm) are at near-resonance with the E_1 gap of InAs at room temperature, and two (496.5 and 457.9nm) are on resonance with the E_1 gap at room temperature and 10K, respectively. As the sampling depth increases, the intensity ratio of the LO and L_- modes decreases, supporting the assertion that the LO

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and L_- modes arise from scattering in the CAR and bulk, respectively. Comparison with data collected on undoped InAs reveals the width of the CAR to be 35 ± 3Å. Some sputter-induced disorder in the first few atomic layers of InAs is evidenced by a slight reduction and broadening of the LO mode relative intensity in the $x(y,y)\bar{x}$ geometry, while no TO mode is seen. [12]

Temperature-dependent Raman spectra of Nb/InAs structures taken through the Nb film are shown in Figure 1. Below T_c the LO phonon scattering intensity, relative to that of the L_- mode, is enhanced by 40%, as determined by lineshape-fitting. This spectral change is reversible upon cycling through T_c, either by changing bath temperature or by changing the surface temperature with laser fluence. This LO-mode enhancement is not seen when scattering on bare InAs, nor is it seen on Nb/InAs when there has not been an *in-situ* etch of the Nb prior to deposition, so that an oxide barrier, ~30Å thick, remains between the Nb and InAs. We therefore conclude that the reversible effects observed in the temperature-dependent Raman spectra require good electrical contact between InAs and Nb.

4. DISCUSSION

Changes in the Raman spectra are not due to superconductivity in the Nb metal alone as they are observed only when Nb is in good electrical contact with InAs. Furthermore, the superconducting gap, less than 1 meV here, can effect neither the visible probing light nor the ~29meV Raman shifts. The increase in LO phonon relative intensity may be attributed to the following four effects occurring in the CAR listed here, and briefly discussed below: 1) Increase in the CAR volume; 2) Increase in the disparity of the electronic properties with the bulk; 3) Increase in the carrier concentration; and 4) Phase correlation of the carriers.

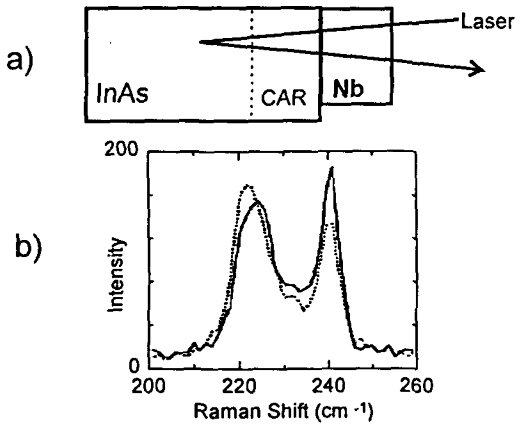

Figure 1. *a) Sketch of experimental geometry. b) Raman spectra of Nb/InAs above and below T_C shown by dotted and full lines, respectively. Below T_C, the relative intensity of the LO mode is increased by ~40%. (After ref. 5,6)*

Since the thickness of the CAR is fixed by the InAs doping level, it is difficult to understand how this width changes upon the T_c crossing. The observation of the LO mode itself is due to the disparity of the electronic properties of the near-surface and bulk regions, and it has been suggested that an increase of this disparity would enhance the LO mode.[12] The proximity effect would increase the CAR conductivity, but how that affects Raman intensity is unclear. Considering 3), charge transport from the Nb into the InAs increases the carrier concentration in the CAR and enhances the Raman cross-section of the LO mode relative to that of the L_-. Below T_c the electrons and holes in the InAs CAR have developed some degree of correlation arising from the Andreev reflection at the interface, thus we consider 4). Assuming a clean interface and taking into account the differences in the effective masses and Fermi momenta of the two materials, the ideal probability for Andreev reflection is 83%. To our knowledge, the effect of a correlated electron plasma on the Raman lattice-mode intensity has not been experimentally or theoretically investigated. Therefore, comparing 3) and 4), it is possible that an increase in the carrier concentration in the CAR can account for the observed effect, but it is also noted that the effect of correlated electrons on Raman cross-section is not known. Therefore, the controversy between Cooper-pair tunneling and reflectionless tunneling has not been settled with the present set of experiments .

5. CONCLUSION

Dramatic changes in the near-surface electronic properties of InAs in intimate electrical contact with superconducting Nb are observed by Raman spectroscopy. This superconducting proximity effect is manifested by the LO phonon mode, associated with the CAR at the interface, exhibiting a marked increase in relative intensity as compared with the L_- coupled plasmon-phonon mode.

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