Photoemission Spectroscopy of the Strong-Coupling Superconducting Transitions in Lead and Niobium

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We study the changes in the electronic structure associated with low-T_c strong-coupling superconducting transitions in Pb and Nb using ultrahigh-resolution (2.3 meV) temperature-dependent (5.3–12.0 K) photoemission spectroscopy. We observe peaks in the density of states on entering the superconducting-phase accompanying gap formation and spectacular redistribution of spectral weight at low energy scales as a function of temperature. The well-known peak-dip feature of the high-T_c cuprates is seen in Pb, making it a characteristic of strong-coupling superconductivity.

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The nature of quasiparticle and collective excitations reveal the mechanism of superconductivity in a system. For the low-T_c superconductors, there exists a clear method to distinguish between the Bardeen-Cooper-Schrieffer (BCS) weak-coupling mechanism as compared to strong-coupling superconductivity. From thermodynamic measurements, BCS theory predicts a value of $2\Delta(0)/k_BT_c = 3.53$, while the strong-coupling superconductors show higher values, where $\Delta(0)$ is the superconducting gap at $T = 0$ (Ref. [1]). The size of the specific heat jump at the transition is also higher for the strong-coupling superconductors than the BCS prediction. Tunneling conductance [2,3] provided the most significant results on crossing into the superconducting phase: a quasiparticle peak and a temperature-dependent gap, features due to phonons and a dip feature at characteristic energy scales. The electron-phonon spectral density $\alpha^2F(\omega)$ was obtained [3] using the Eliashberg equations. The tunneling results provided a detailed comparison to the theory of strong electron-phonon coupling mediated superconductivity in metals [4]. Phonon features and a dip in the tunneling conductance are consistent with strong-coupling theory but not with BCS theory. In this work, we study the form of angle-integrated photoemission spectra for strong-coupling superconductors Pb and Nb, as a function of temperature (5.3 to 12.0 K) across the superconducting transition. Photoemission is a one-electron removal process and is thus analogous to superconductor-insulator-normal (S-I-N) tunneling as a probe of the single-particle density of states (DOS), normalized to their respective transition probabilities. Using ultrahigh resolution (2.3 meV), we show that angle-integrated photoemission spectroscopy provides a one to one correspondence to features observed in S-I-N tunneling: We observe a peak in the DOS on entering the superconducting phase, bulk phonon features, a dip feature, and a temperature-dependent gap. The peak and dip feature is similar to high-T_c cuprates, making it a characteristic of strong-coupling superconductivity. The present study makes photoemission spectroscopy a reliable technique to probe changes in single-particle DOS even at very low energy scales.

The present work has been carried out on a new photoemission spectrometer built at ISSP, using a monochromatic He I source (GAMMADATA), Scienta SES2002 analyzer, and a newly designed thermally shielded sample holder with a flowing liquid He cryostat. The sample was mounted on this thermally shielded sample holder, and the sample temperature was measured using a calibrated silicon diode sensor to an accuracy of ±0.5 K. Clean surfaces were obtained by scraping the sample surfaces *in situ* with a diamond file. The total experimental resolution is 2.3 meV as determined for the Fermi edge of a gold film measured at 5.3 K. The position of the Fermi level is accurate to better than ±0.05 meV. All spectra shown are an average of at least 300 scans, with temperature-dependent changes reproducibly obtained on thermal cycling.

Figure 1 shows the photoemission spectra of (a) Pb and (b) Nb, measured at 5.3 and 12.0 K, across the superconducting transition at $T_c = 7.19$ and 9.26 K, respectively. For Pb, we observe a sharp peak at 2.5 meV binding energy in the spectrum at 5.3 K, followed by a weaker peak at about 9 meV and a dip between 10–15 meV. The opening of a superconducting gap leads to a shift in the leading edge of the spectrum and redistribution of spectral weight up to 15 meV as compared to the normal-phase spectrum. Similarly, Fig. 1(b) shows gap formation in the Nb spectrum, a sharp peak at 2.7 meV, and redistribution of spectral weight up to about 15 meV. Thus, in both cases, the spectral weight is conserved across the superconducting transitions over an energy scale of about 15 meV, which is much larger than the gap energy or T_c. We estimate values of the gap using a Dynes' function [5] fit (Fig. 2) to the peak in the DOS, given by $N(E,\Gamma) = (E - i\Gamma)/[(E - i\Gamma)^2 - \Delta^2)]^{1/2}$. The real part gives us the measured spectra $N(E)$, Δ is the gap, and Γ is a measure of the thermal broadening due to finite lifetime of

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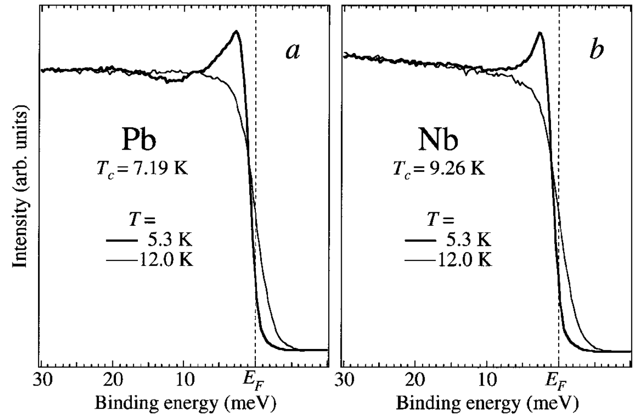

FIG. 1. Ultrahigh-resolution photoemission spectra of (a) Pb and (b) Nb, measured at 5.3 K (superconducting phase) and 12.0 K (normal phase). Redistribution of spectral weight and the opening of superconducting gaps are observed for Pb and Nb.

the quasiparticles at the gap edge. We obtain values of Δ (5.3 K) = 1.20 meV and a Γ = 0.08 meV for Pb, and Δ (5.3 K) = 1.35 meV and a Γ = 0.10 meV for Nb. Using the measured values of Δ (5.3 K) and the known dependence of the reduced energy gap $\Delta(T)/\Delta(0)$ versus reduced temperature (T/T_c) from strong-coupling theory [4] (which is also very similar to the BCS weak-coupling result as well as other experiments), we obtain values of $2\Delta(0)/k_B T_c$ for Pb to be 4.9 and for Nb to be 3.7. These values of $2\Delta(0)/k_B T_c$ are in good agreement with values known from thermodynamic measurements—4.5 for Pb and 3.8 for Nb (Ref. [1]). In reference to measurement of the superconducting gap by photoemission spectroscopy, it has been possible to date only for the high-T_c superconductors. In fact, high-resolution angle-integrated and angle-resolved photoemission spectroscopy (ARPES) has provided many important results in the study of high-T_c superconductivity. Some of the most important results are the measurement of the superconducting gap with a relatively large value of $2\Delta(0)/k_B T_c$ (~5–8) [6–8] motivating strong coupling as a possible mechanism of high-T_c superconductivity, $d_{x^2-y^2}$ symmetry of the superconducting gap [9,10], and existence of a pseudogap above T_c (Refs. [11–13]).

The Dynes' function fit deviates from experiment at binding energies beyond the peak in the DOS, particularly for Pb. In Pb, at 5.3 K, we see in Fig. 1(a) (i) a weak feature at about 9 meV, (ii) the peak in the DOS is itself asymmetrically broadened on the higher binding energy side (4–6 meV), and (iii) a dip at 10–15 meV. This is more clearly seen when we divide the superconducting-phase spectrum by the normal-phase DOS (obtained from the normal-phase spectrum divided by a corresponding Fermi function), as is shown in Fig. 3. It is known from neutron scattering studies [14] and from a strong-coupling analysis of the tunneling spectra [2–4] that Pb exhibits a transverse phonon at 4.4 meV and a longitudinal phonon at 8.5 meV, and we attribute the features seen in photoemission also to the same origin. For Nb, we do not see a phonon feature, expected at about 15 meV (Ref. [15]), though the spectral weight redistribution occurs with higher intensity in the superconducting phase up to about 15 meV binding energy. We also note that the spectral weight redistribution in Nb is not as spectacular as in the Pb spectrum (see Fig. 1). We believe this is due to the weaker coupling in Nb compared to Pb, which is well-known from thermodynamic measurements [1].

Though this is the first observation of phonon features upon entering into the superconducting phase of a low-T_c strong-coupling system using photoemission spectroscopy, discrete loss features due to excitation of phonons have

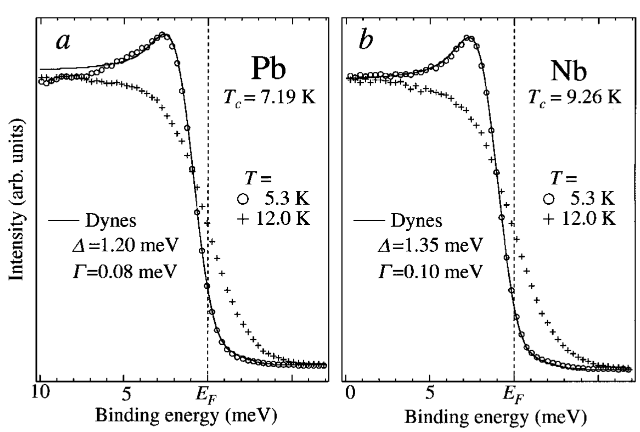

FIG. 2. Dynes' function fits to the peak in the DOS for (a) Pb and (b) Nb are used to estimate the superconducting-gap values.

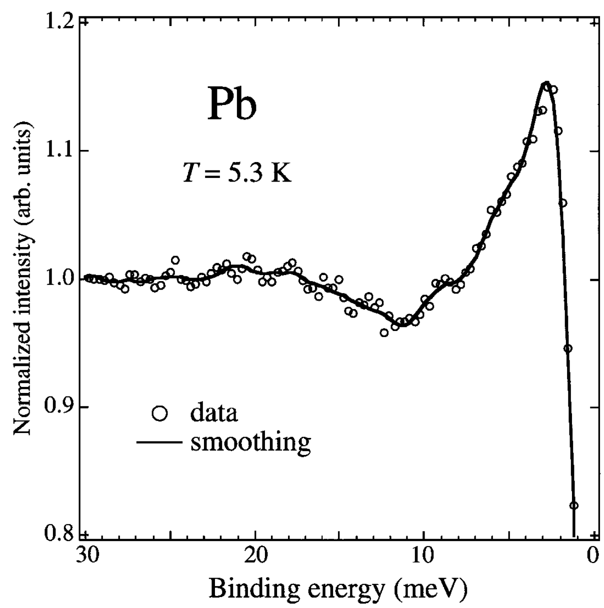

FIG. 3. Photoemission intensity I(5.3 K)/DOS(12.0 K) for Pb, plotted in order to see the details in the superconducting-phase spectra in analogy to tunneling conductance experiments.

been observed recently in photoemission spectra of C_{60}^- clusters [16]. In that study, electron-phonon coupling constants were derived for C_{60}^- and used to support an electron-phonon mechanism for superconductivity in the fullerides. Further, for the cuprate superconductors, strong coupling has been established between the collective resonance mode observed at the wave vector (π, π) by neutron scattering [17,18] and the resonance peak-dip-hump line shape observed at $(\pi, 0)$ in angle-resolved photoemission spectroscopy [19,20]. The origin of the peak-dip-hump structure in the cuprates is due to scattering of magnetic excitations by electrons [21]. In contrast, the strong coupling in low-T_c superconductors like Pb is between electrons and phonons. Our results show that the peak-dip structure is also observed in a low-T_c system and is thus a characteristic of strong-coupling superconducting transitions. The present study gives a reference to compare the mechanism of superconductivity and the changes in the density of states for the low-T_c systems *vis-à-vis* the high-T_c superconductors using photoemission spectroscopy.

In an attempt to study the temperature dependence of the density of states in the superconducting phase, we measured the photoemission spectra of Nb from 5.3 to 10.3 K (just above $T_c = 9.26$ K), as shown in Fig. 4(a). The spectra do show a clear evolution in the superconducting phase as a function of temperature, with a systematic shift of the leading edge and increasing spectral weight transferred into the peak in the DOS on lowering temperature. In the inset of Fig. 4(a), we plot the shift in the midpoint of the leading edge as a measure of the changing superconducting gap versus reduced temperature (T/T_c). The temperature dependence between 5.3 K and $T_c = 9.26$ K confirms the theoretically expected dependence for strong-coupling superconductivity, which is similar to the BCS result as mentioned earlier. Following Norman *et al.* [22], we plot symmetrized spectra of Nb in Fig. 4(b), so as to see the changes occurring in the density of states as a function of temperature. We note that symmetrization is valid for the case of particle-hole symmetry measured in ARPES spectra at low energy scales and $k = k_F$. Here, since we are investigating changes in the DOS of Nb, the normal phase of which is well approximated by a constant DOS within 10 meV of E_F indicating particle-hole symmetry, we find symmetrization is meaningful. The resulting spectra show the gap opening progressively with depletion of spectral weight around the Fermi level for decreasing temperatures, quite like tunneling conductance spectra.

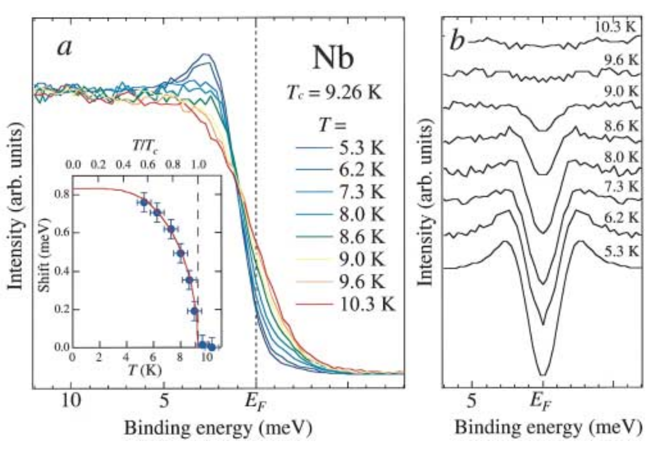

FIG. 4 (color). (a) Temperature-dependent photoemission spectra of Nb (5.3–10.3 K), measured to investigate the changes in the density of states in the superconducting phase. Inset shows a plot of the shift in the midpoint of the leading edge versus reduced temperature, confirming the temperature-dependent gap. The red curve in the inset corresponds to a result of temperature dependence of the gap with $\Delta(0) = 0.83$ meV based on the strong-coupling theory [4]. (b) Symmetrization analysis of the spectra shown in (a), indicating a systematic depletion in spectral weight about the Fermi level due to an increasing gap on lowering the temperature.

In conclusion, using ultrahigh-resolution photoemission spectroscopy as a probe to study the strong-coupling superconducting transitions in low-T_c Pb and Nb, we obtain the following results: The spectra exhibit a peak due to pileup in the DOS and discrete loss features due to phonons in the superconducting phase, a dip feature, and a temperature-dependent gap with systematic redistribution of spectral weight. Photoemission spectroscopy is thus shown to be a reliable technique to probe single-particle density of states even at very low energy scales. The present study opens up possibilities for studying low temperature phase transitions, and, in particular, superconductivity, and their relation with changes in the density of states measured using photoemission spectroscopy at very low energy scales.

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