Tunneling Spectroscopy in AlNiCo Decagonal Quasicrystals

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Tunneling spectroscopy of $Al_{73}Ni_{17}Co_{10}$ decagonal single quasicrystals has been measured at ultralow temperatures. In addition to the wide pseudogap observed in previous photoemission experiments, very rich fine structures in the density of states around the Fermi energy were discovered. The fine structures strongly depend on the applied magnetic field and disappear above ~4 T. The zero-bias resistance of the junction varies with magnetic fields in a complicated way, which is suggested to be related to the hierarchy structure of quasicrystals. A puzzling hysteretic phenomenon was observed in some bias ranges in zero magnetic field, the origin of which needs to be clarified in the future. [S0031-9007(99)08419-7]

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The effect of quasiperiodic order on electronic properties is one of the central problems in the study of quasicrystals. Two types of pseudogaps in electronic structures of quasicrystals are discussed in the literature. One is a Hume-Rothery-like pseudogap, which is related to Brillouin scattering. This pseudogap, of the order of 1 eV, has been predicted by band structure calculations, suggested by specific heat measurements, and observed directly by x-ray emission and photoemission spectroscopy [1]. The wide pseudogap, mainly related to the complex local atomic configuration in first approximation, is responsible for the stability of quasicrystals.

The other type of pseudogap, depending on the coherence length of the long range quasicrystalline order, is expected to have fine and rich structures. Linear muffin-tin orbital (LMTO) calculations of crystalline approximants reveal very spiky structures in the density of states (DOS) [2,3], i.e., there are many peaks and valleys of small width superimposed on the wide pseudogap. There are indications that these spiky features could be associated with the confinement of electrons in clusters [4], and the more clusters are involved, the more spiky is the DOS. It is obvious that the fine structures of the DOS are more important and more interesting to the physics of quasicrystals. However, since the long range quasicrystalline order energetically makes only a small perturbation to the main pseudogap, and the longer the coherence length, the richer the structure of the energy spectrum, to observe these fine structures one expects to need an energy resolution beyond which could be reached by, say, photoemission spectroscopy. Low temperature tunneling spectroscopy is powerful in this respect. In the past few years, tunneling spectroscopy has been used to study the pseudogap for icosahedral quasicrystals [5,6]. Although a well-defined pseudogap was observed, no spiky peaks were found for the i-phase samples. Before one concludes a violation of these measurements to the band structure calculations, we notice that all of these investigations were carried out on thin film samples [5] or ribbons [6]. Since a tunneling experiment is sensitive to the surface condition of the sample, and the perfectness of the quasicrystalline order could be crucial to the fine structure of DOS, it would not be unreasonable that these experiments failed in detecting the spiky structures. Furthermore, recent experiments show that even the fresh-cleaved surface of AlPdMn icosahedral single quasicrystals is rough [7] and no STM atomic resolution could be obtained [8]. Atomic resolution was obtained on a tenfold surface of decagonal AlCuCo quasicrystals, and full decagonal symmetry was observed [9]. Therefore, it would be attracting and revealing to investigate the tunneling spectrum of the decagonal samples.

This Letter reports the first tunneling spectroscopy study on decagonal single quasicrystals at ultralow temperatures (down to 25 mK). Very rich structures, characterized by a zero-bias peak in DOS and many anomalies in the spectra, were observed. The tunneling spectra can be changed by an applied magnetic field and evolved to a single pseudogap as the magnetic field increases.

The $Al_{73}Ni_{17}Co_{10}$ decagonal single quasicrystals were grown by a method similar to that described in Ref. [10]. The high quality of the samples were checked in many ways [11]. The samples exhibit a columnar decagonal prismatic morphology with the column axis parallel to the periodic direction. One end of the columnar sample was plugged into a pinhole of a phosphorous bronze spring platelet. The other end, with a fresh-broken surface approximately perpendicular to the tenfold axis, was softly touched on an Al film of ~50 nm in thickness deposited on a glass substrate by magnetron sputtering. The pressure between the sample tip and the Al film could be adjusted by a tiny screw. The Al film was chosen because the oxide layer on top of the film is self-limiting in the tunneling thickness range and is uniform and pinhole-free. It was proved that stable tunneling junctions with junction resistances ranging from several ten to several hundred ohms could often be made in this way. The experiment was conducted in a dilution refrigerator with a superconducting magnet. The magnetic field was

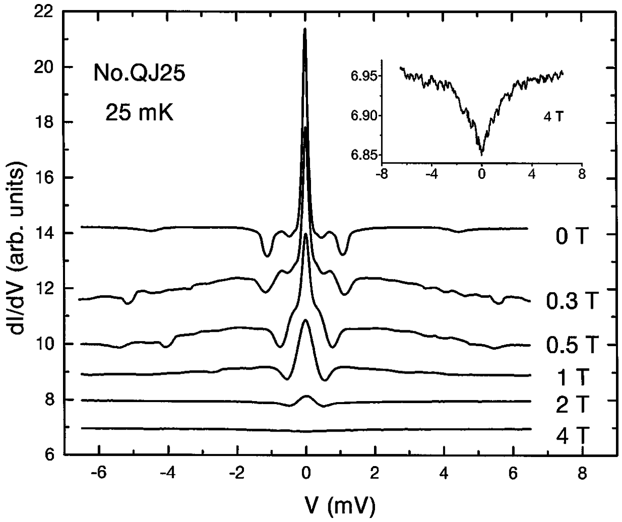

FIG. 1. Tunneling spectra of a decagonal AlNiCo single quasicrystal, No. QJ25, with an Al film as a counterelectrode at 25 mK. The curves have been shifted in line successively by 1 unit upward from that in 4 T for clarity. Fine structures can be seen in low magnetic fields and they disappear above 4 T. Inset is an enlarged plot of the curve in 4 T, which demonstrates the existence of a pseudogap.

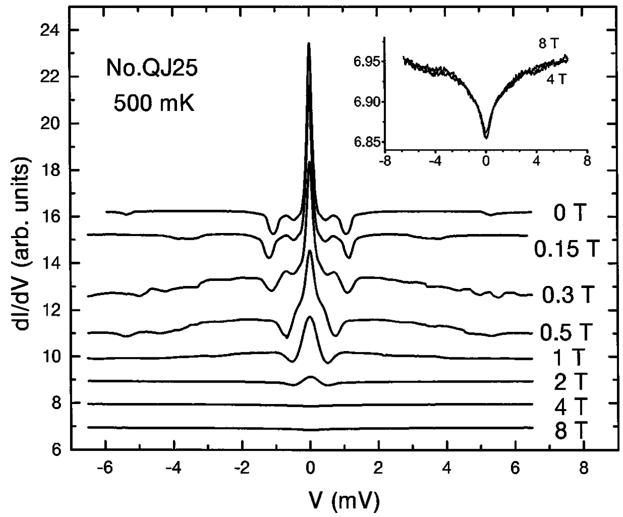

FIG. 2. Tunneling spectra of a decagonal AlNiCo single quasicrystal, No. QJ25, with an Al film as a counterelectrode at 500 mK. The curves have been shifted in line successively by 1 unit upward from that in 8 T for clarity. Fine structures can be seen in low magnetic fields and disappear above 4 T. Inset is enlarged plots of the curves in 4 and 8 T (without shift). The tunneling spectra do not change significantly from those at 25 mK.

parallel to the Al film and also to the quasicrystalline plane of the AlNiCo samples. The dI/dV vs V was measured by slowly sweeping the dc bias modulated with a small ac signal (1.097 kHz, 0.1 mV) in standard four-probe configuration.

Figures 1 and 2 show typical tunneling spectra obtained at 25 and 500 mK, respectively, for one (No. QJ25) of the four measured samples. Fine structures can be clearly seen in the spectra. The structures do not change significantly up to 500 mK, and are reproducible for the same sample in several runs of measurements, but the details of the fine structures are sample dependent. We believe that this sample dependence could be closely related to the perfectness of the quasicrystalline order and the composition fluctuation of the samples. As an example, the spiky structures in zero magnetic field for another sample, No. QJ21, are somewhat different (Fig. 3). In spite of the details of the fine structures, two main features are common to the tunneling spectra of all of the measured samples. First, there is always a zero-bias peak in the absence of magnetic fields. Second, the fine structures including the zero-bias peak, change with the applied magnetic field and finally disappear when the field is high enough, leaving a simple, narrow pseudogap at the Fermi energy (Figs. 1 and 2, insets).

Before discussing their behavior in detail, we must be sure that the tunneling spectra observed here are intrinsic to quasicrystals. We notice that a zero-bias peak in conductance has also been seen previously in two types of systems: tunneling junctions with superconductors involved [12], often understood with the picture of Andreev reflections, and junctions with magnetic impurities involved, understood by Kondo-assisted tunneling [13,14].

Since in our case an Al film was used as the counterelectrode, forming a quasicrystal-Al_2O_3-superconductor junction, it would be natural to think that the superconductivity of Al could be responsible for the fine structures observed here. This possibility can be completely ruled out for the following reasons. First, in the superconductivity-related tunneling spectra, all structures are restricted within the superconducting energy gap 2Δ [12]. The transition temperature T_c for our 50 nm thick

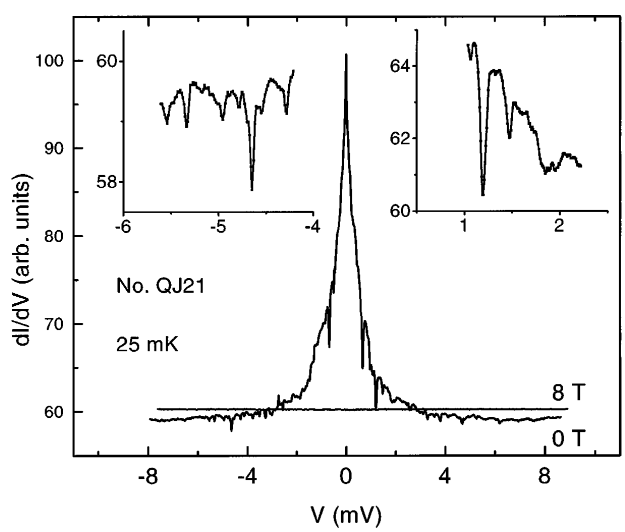

FIG. 3. Tunneling spectra of a decagonal AlNiCo single quasicrystal, No. QJ21, with an Al film as a counterelectrode at 25 mK. Fine structures of the spectrum in zero magnetic field can be clearly seen in comparison with the smooth curve observed in 8 T. Parts of the zero-field tunneling spectrum are enlarged in the insets.

films is close to the bulk value, leading to 2Δ less that 0.4 meV. Therefore, the fine structures we observed are dominantly far outside 2Δ. The zero-bias peak even covers a width about 10 times 2Δ for the sample, No. QJ21 (Fig. 3). Second, the peak width does not shrink with the magnetic field H up to 2 T, violating what should be expected for superconductors. Third, the peak height as a function of H shows some structures [see below, (Fig. 4)] which cannot be explained by Andreev reflections. The suppression of the superconducting gap of Al in the spectra is due to a reminiscent pinning magnetic field about 0.02 T, which, for convenience, is neglected when discussing the behavior of quasicrystal samples.

The possibility that the rich structures of our tunneling spectra are caused by Kondo-assisted tunneling can be excluded. The latter promises only a single peak at Fermi energy when $H = 0$, and the peak should split into two peaks gradually departing the zero-bias with the increase of H [13,14]. In contrast, besides the rich structures, the position of our zero-bias peak does not change with H, while the adjacent structures show nonmonotonous dependence on H, as can be seen in Figs. 1 and 2.

From the above discussion, we believe that the observed tunneling spectra are intrinsic to the AlNiCo decagonal quasicrystals. Now we show the peculiar behavior of the spectra in such a junction.

According to the standard theory of electron tunneling spectroscopy [13], the measured spectra in Fig. 1 is directly related to the DOS of quasicrystals. Therefore, the zero-bias peak in low magnetic fields means that the DOS of the decagonal quasicrystals has a maximum at Fermi level E_F. The appearance of this peak and a small hump in a wider range (~40 meV) certainly does not affect the

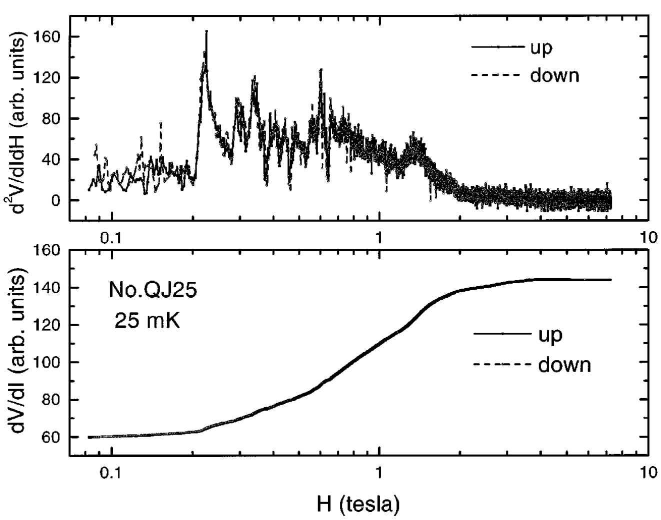

FIG. 4. Zero-bias tunneling magnetoresistance (lower panel) and its derivative (upper panel) of the junction with the decagonal AlNiCo single quasicrystal, No. QJ25, and with an Al film as a counterelectrode at 25 mK. The structures in the curves, well reproduced in the up and down runs of the magnetic field, are approximately evenly spaced in the semilogarithmic plot.

stability of the system, since, as we mentioned above, these structures make only a perturbation compared to the much wider pseudogap (~1 eV), which is determined mainly by the local bond configurations of the material. The existence of a zero-bias peak is supported by theoretical LMTO calculations on a model approximant of the decagonal quasicrystal AlCuCo [3]. The calculations show that the Fermi level is indeed near a sharp peak of the spiky DOS.

A fascinating feature of the tunneling spectra is their strong H dependence (Fig. 1). The DOS, as a function of magnetic fields of two- and three-dimensional quasicrystalline model systems, has recently been calculated in a tight-binding description [15]. It shows that there is a trend of the eigenstates to get a more uniform distribution over the whole energy band with increasing magnetic fields. This is qualitatively consistent with the spectra in Fig. 1. The fine structures become undetectable when the field is higher than 4 T. We may further demonstrate the peculiar H dependence by the measurement of zero-bias dV/dI as a function of H. The result is shown in the lower panel of Fig. 4. Some structures immediately emerge when we differentiate the curve with respect to H (Fig. 4, upper panel). The structures were well reproduced as we swept the magnetic field slowly up and down. It is interesting to note that the main structures are approximately evenly spaced when we plot the magnetic field axis in a logarithmic scale. This implies that a rapid change of the tunneling resistance occurs each time when the magnetic field increases exponentially. Increasing the field means decreasing the area containing a magnetic flux quanta. Since the magnetic field is applied parallel to the tenfold surface of the decagonal quasicrystal, we may assume a H-independent characteristic length along the tenfold axis, and then the exponentially increased fields correspond to an exponentially decreased length scale in the quasicrystalline plane. So it is suggestive that the tunneling magnetoresistance reveals the self-similar properties of the quasicrystalline structure, which has hierarchy scales. We can further make a rough estimation about the length scales in the quasicrystalline plane. Choosing the characteristic length along the tenfold axis as 200 nm, which is the correlation length measured from x-ray diffraction [9], we find that 0.22 T, the location of the first high peak in Fig. 4, corresponds to 47 nm and 4 T, at which fine structures of the tunneling spectra disappear (see Fig. 1), corresponds to 2.1 nm. To further clarify the field dependence of the spectra, the zero-bias magnetoresistance of the junction needs to be measured as a function of the directions of H with respect to the quasicrystalline plane.

The most puzzling feature was observed when we extended the measurements to high bias (Fig. 5). A reproducible hysteresis was observed in ranges of $\sim -23 - -7$ mV and $\sim 7-23$ mV. The hysteresis disappeared together with the fine structures at high H. To our knowledge, there was no report on a similar hysteresis

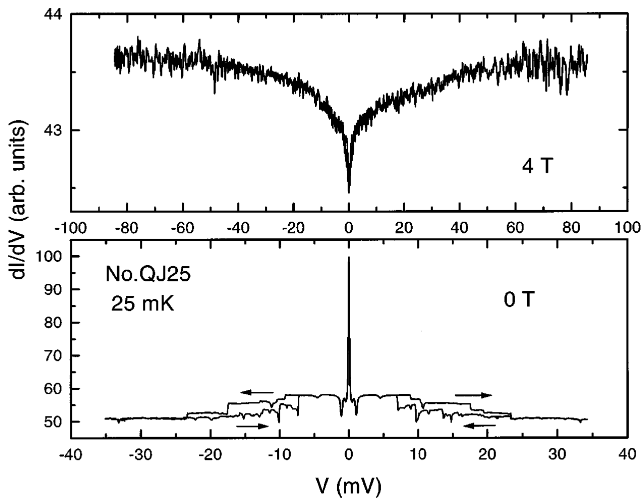

FIG. 5. High-bias tunneling spectra of a decagonal AlNiCo single quasicrystal, No. QJ25, with an Al film as a counterelectrode at 25 mK. The spectra show a puzzling hysteresis in some bias ranges in zero magnetic field (lower panel). The hysteresis disappears together with the fine structures in the field of 4 T (upper panel).

in tunneling spectra. The hysteresis is serious because it challenges the DOS explanation of the data. To keep the DOS explanation workable, some amendments are needed. A possible one is to assume that the long range coherence of the quasicrystalline order at the junction can be enhanced in some way by the electric field in the junction area. This enhancement occurs when the field is stronger than ± 23 mV, and collapses only when the field decreases to ± 7 mV, resulting in a reversible metastable change. An alternative explanation is that the mismatch between the AlNiCo quasicrystal and its counterelectrode in both energy spectra and dynamical properties of electrons leads to the hysteresis. Other effects may be involved in high bias spectra. Further investigations are needed for this phenomenon.

In summary, we have measured the tunneling spectra of decagonal AlNiCo single quasicrystals at ultralow temperatures. Complicated tunneling spectra have been observed, which are reminiscent of the spiky structures in DOS expected by model calculations. The dependence of the spectra on the magnetic field is qualitatively consistent with recent theoretical results. The zero-bias behavior in magnetic fields shows anomalies which indicates hierarchy structures. A puzzling hysteresis was observed in some bias ranges at $H = 0$. All of the above findings call for further experimental and theoretical efforts.

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