Point Contact Properties of $R_3Pd_{20}X_6$
(R = La, Ce; X = Si, Ge) Cage Compounds

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Point-contact spectroscopy measurements have been performed on the $R_3Pd_{20}X_6$ (R = La, Ce; X = Si, Ge) cage compounds. In case of La the characteristic phonon energies have been obtained in agreement with that of the Raman scattering. In $Ce_3Pd_{20}Si_6$ we have observed asymmetric behaviour of $dV/dI(V)$ dependences which was observed in non-Fermi liquid compound. In $Ce_3Pd_{20}Ge_6$ we have observed maxima at crystalline electric field energies, influenced by magnetic field. This is connected with quadrupolar ordering transition.

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1. Introduction

$RE_3Pd_{20}X_6$ is a series of rare-earth (RE) based isostructural intermetallic compounds that form with either X = Si or Ge, crystallizing in cubic structure [1]. The structure of the unit cell in $RE_3Pd_{20}X_6$ is amenable to a cage — or clathrate — like description due to the structural environment of the rare-earth atoms [2]. This kind of compound forms an interesting class of materials, whose magnetic properties reflect the existence of two distinct rare-earth atom sites of cubic point symmetry. Different types of long-range order (magnetic, quadrupolar) observed at low temperatures have been ascribed to a separate ordering of the two sublattices [2]. These compounds are also interesting as potential candidates for thermoelectric applications. They are characterized by strong involvement of optical phononic modes due to anharmonic thermal vibrations of rare earth atoms situated on the tetrahedral symmetry 4a-site cage. These rattling modes have been studied by ultrasonic attenuation at low temperatures (e.g. [2]) and in case of La by the Raman scattering [3].

2. Method

In order to see directly the characteristic energies of such modes we have used the point contact (PC) spectroscopy technique [4], which enables the direct observation of the electron–quasiparticle interaction. The method is based on the measurements of the second derivative of I–V characteristics of metallic PCs, which is directly proportional to the electron–quasiparticle interaction function of studied metals. The comparison of $La_3Pd_{20}Si_6$ PC spectra containing only the information about electron–phonon interaction with Ce compounds shows the magnetic contribution to the properties of these compounds.

3. Experimental

$La_3Pd_{20}Si_6$, $Ce_3Pd_{20}Si_6$ and $Ce_3Pd_{20}Ge_6$ samples have been prepared by congruent arc-melting of the stoichiometric quantities of the constituent elements in ultra-high purity argon. Elemental purities were (wt%) Ce, La and Pd (99.99), Si and Ge (99.9999). The point contacts were made at liquid helium temperatures by touching a Cu or Pt needle (heterocontacts arrangements) into the polished surface of the $RE_3Pd_{20}X_6$ samples. The derivatives $d^2V/dI^2(V)$ and $dV/dI(V)$ of the I–V characteristics were measured using a standard PC technique [4] in the temperature range from 1.5 K to 10 K.

In order to obtain the PC spectra of good quality we have measured in the heterocontact arrangement where one electrode is the metal with a well-known PC spectrum like Cu or Pt and the second electrode is the metal under study. However, the contribution of one metal to the heterocontact PC spectrum is indirectly proportional to the Fermi velocity of this metal [4]. We could expect different Fermi velocities of studied compounds and therefore, we could expect the contribution from $RE_3Pd_{20}X_6$ compounds mainly.

4. Results and discussion

In the case of $La_3Pd_{20}Si_6$, which is characterized by electron–phonon interaction only, we have observed the "classical" metallic point contact spectra $d^2V/dI^2(V)$, presented in Fig. 1 for different PC resistivities. PC spectra are shifted for the clarity on y-axis. In case of heterocontacts with Pt, two maxima were observed: one at voltages between about 9.2 mV and 10.5 mV and another small maximum at about 14 mV. The energy positions of these maxima agree well with the maxima at characteristic phonon energies and rattling mode, observed in the Raman spectra [3]. In case of heterocontact with Cu, the

structure was less pronounced. However, except of maximum at 9.2–10.5 mV, the small maxima at 5 mV and at 7 mV were observed. The 14 mV maximum found using Pt heterocontacts was also found to be absent. Moreover, a large shoulder between 15 mV and 20 mV was observed which is characteristic for Cu phonon energies [4].
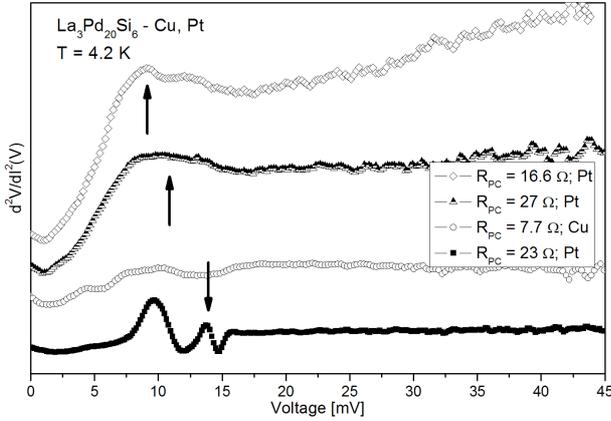
Fig. 1. Characteristic PC spectra $d^2V/dI^2(V)$ of $La_3Pd_{20}Si_6$–Cu, Pt heterocontacts at 4.2 K in spectroscopic regime.

By contrast, in the magnetic $Ce_3Pd_{20}Si_6$ compound a much more pronounced behaviour was found within our Pt heterocontact measurements. Already the first derivative $dV/dI(V)$ shows a sharp maximum in the vicinity of zero bias. In Fig. 2 such characteristic behaviour of PC dependences $dV/dI(V)$ at 1.5 K as function of an applied magnetic field is shown. However, the maximum is asymmetric and it is placed in negative voltage polarity at about –1 mV. With increasing magnetic field it shifts to higher voltages (–2.5 mV at 6 T). This type of asymmetry was for the first time observed in the PC dependences of heterocontacts between non-Fermi liquid compounds in the system $YbCu_{5-x}Al_x$ and Pt or Cu [5]. This is in agreement with the observation of possible quantum critical behaviour in $Ce_3Pd_{20}Si_6$ [6], induced by magnetic field. With increasing temperature the asymmetry remains the same and it does not depend on the magnetic field up to 6 T.

Substitution of Si by Ge yields to the formation of the Kondo lattice in compound $Ce_3Pd_{20}Ge_6$ [1]. This changes also the character of PC spectra. In Fig. 3 the characteristic PC spectra $d^2V/dI^2(V)$ of heterocontact between $Ce_3Pd_{20}Ge_6$ and Pt as function of an applied magnetic field are shown. At zero applied magnetic field we observed three small maxima: at 1.5 mV, 4.1 mV and broader at about 5.2 mV. The last two values are in good agreement with the crystal electric field (CEF) observation by neutron inelastic scattering [7]. With an applied magnetic field 3 T and 6 T only one more intense peak at 1.5 mV appears. It is followed by large shoulder at about 20 mV, which is of phononic origin. This behaviour confirms the magnetic origin of low energy peaks. Therefore,
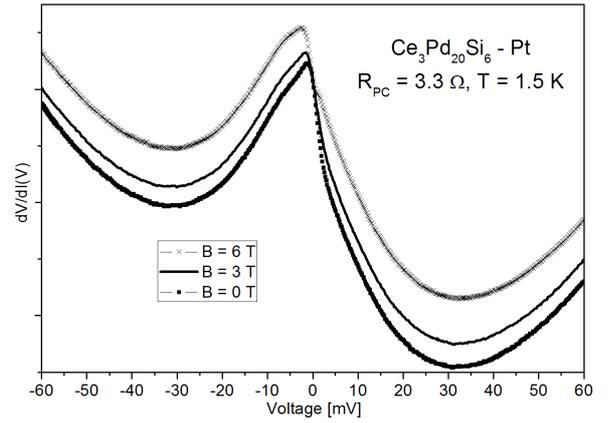
Fig. 2. Magnetic field dependence of PC $dV/dI(V)$ dependences at 1.5 K for heterocontact $Ce_3Pd_{20}Si_6$–Pt.

we suppose that first peak has its origin in CEF excitations and all the resolved lower energy anomalies are connected with allowed CEF excitation from non-singlet ground state to the excited state.
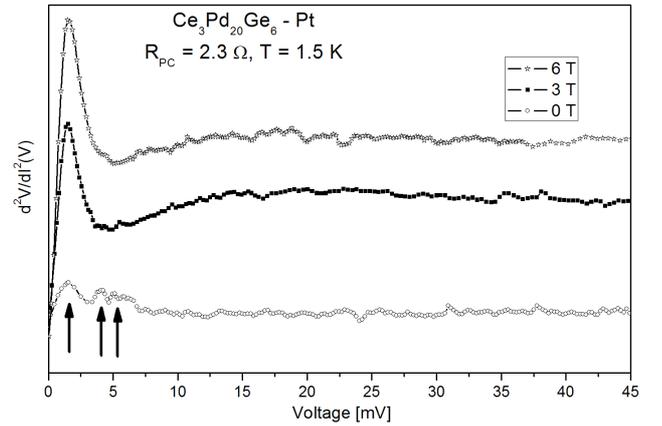
Fig. 3. Magnetic field dependence of the characteristic PC spectra $d^2V/dI^2(V)$ of $Ce_3Pd_{20}Ge_6$–Pt heterocontact at 1.5 K in spectroscopic regime.

Magnetic field is known to suppress the Kondo behaviour in $Ce_3Pd_{20}Ge_6$ by arresting spin degrees of freedom associated with incoherent spin fluctuations [8]. Taking into account the observation of quadrupolar ordering in phase diagram of $Ce_3Pd_{20}Ge_6$ [9], the quadrupolar phase transition is present at 1.5 K above magnetic field 3 T. Therefore, we suppose the change of peaks character (three less intense in zero field and one sharp with higher intensity in field) is connected with this phase transition.

5. Conclusions

In conclusion, we have measured the PC dependences of heterocontacts between $La_3Pd_{20}Si_6$, $Ce_3Pd_{20}Si_6$ and

$Ce_3Pd_{20}Ge_6$ and Pt or Cu. In $La_3Pd_{20}Si_6$ we have observed the electron–phonon interaction with characteristic phonon energies. In $Ce_3Pd_{20}Si_6$ we have observed asymmetric behaviour of $dV/dI(V)$ dependences which was observed in non-Fermi liquid compound $YbCu_{5-x}Al_x$ too. In $Ce_3Pd_{20}Ge_6$ we have observed maxima located at previously established CEF energies, which are influenced by magnetic field. This is connected with quadrupolar ordering transition.

Acknowledgments

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