Observation of electron interaction with local vibrations in metallic microcontacts

Yu. G. Naĭdyuk, N. A. Chernoplekov, Yu. L. Shitikov, O. I. Shklyarevskiĭ, and I. K. Yanson

I. V. Kurchatov Institute of Atomic Energy

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Interaction between conduction electrons and local vibrations is observed in Cu-Be α solutions by the microcontact spectroscopy technique. The density dependences of the local-vibration parameters (intensity and energy width) are studied and compared with data on cold-neutron elastic scattering. The efficiency of electron scattering by the impurity-center vibrations is found to be the same as that for scattering by longitudinal acoustic phonons of the matrix. The much larger straggling of the local vibrations observed in the microcontact spectra can be attributed to distinguishing properties of the method employed.

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Study of nonlinear effects in the electric conductivity of a point contact between two normal metals yields direct information on the mechanisms whereby conduction electrons are scattered (see the review[1]). This method, named microcontact (MC) spectroscopy, was used to study in detail electron-phonon interactions (EPI) in many metals,[1,2] electromagnetic interaction,[3] and scattering of electrons by magnetic impurities (the Kondo effect).[4,5]

It is usually assumed that the mean free path l of the electrons, defined by the relation

$$l^{-1}=l_i^{-1}+l_e^{-1}, \quad (1)$$

where l_i and l_e are respectively the mean free paths with respect to momentum and energy relaxation, is much larger than the microcontact diameter d. This condition is satisfied by metals of moderate purity, having an impurity mean free path $l_i \gtrsim 10^{-4}$ cm. However, as shown in Ref. 6, the region of applicability of MC spectroscopy is in fact larger and can be extended to include alloys with small impurity mean free path $l_i \ll d$ under the condition that the energy relaxation length l_e is so large that the diffusion energy-relaxation length $\Lambda_e \approx l_i l_e/3)^{1/2}$ remains large compared with d. Microcontact spectra of a Cu-Ni alloy (disordered solid solution containing up to 20% Ni was investigated experimentally in Ref. 7). It was observed that besides the broadening of the spectral bands and the change of their relative intensity, a shift takes place in the maximum due to the interaction of the electrons with the transverse acoustic phonons (of the TA maximum), towards large energies; this shift corresponds qualitatively to the expected increased rigidity of the lattice with increasing concentration of the Ni atoms. Particular interest, however, attaches to the study of impurity systems in which new vibrational modes (e.g., local or quasilocal vibrations) can appear. It is not clear here beforehand whether the band broadening due to the impurity scattering of the electrons will lead to a smearling large enough to make impossible distinctly localized singularities.

We present in this communication the results of the first observation, in MC spectra, of singularities due to the interaction of conduction electrons with local vibrations caused by the admixture of "light" Be atoms in a lattice of "heavy" Cu atoms. We have investigated Cu-Be alloys with Be concentrations 0.5, 0.9, 2.7, and 4.2 at.%, on which neutron measurements were made[8] of the phonon spectra. The samples in the form of right-angle prisms measuring $1\times1\times10$ mm were bright-dipped in a 60% solution of H_3PO_4 and mounted in a special device for microdisplacements and placed in a cryostat, where a point contact was produced by the method described in Ref. 9 after cooling the system to helium temperatures.

The microcontact spectrum is the second derivative of thecurrent-voltagecharacteristic,$d^2V/dI^2/=f(V)$,which was automatically plotted with an x–y recorder as a function of the voltage applied to the contact.

Figure 1a shows the MC spectra of Cu and Be single crystals. They are proportional to the EPI functions G (eV) of

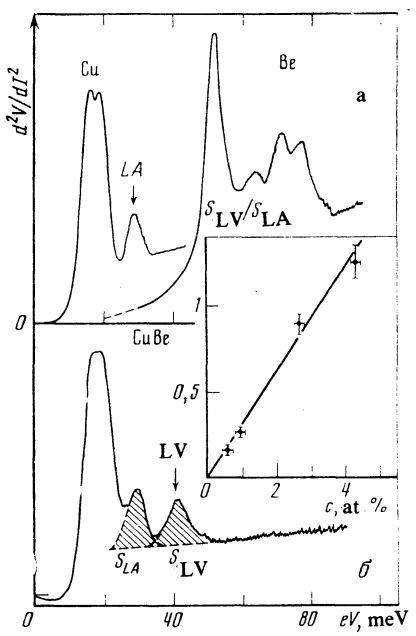

FIG. 1. Comparison of microcontact spectra of copper, beryllium (a) and of a disordered α-solution of Be (2.7 at.%) in a copper matrix (b). Inset—dependence of the relative intensity of the local vibration on the Be impurity concentration.

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these metals, which in turn are connected with the densities of the phonon states of copper and beryllium $F(eV)$ by the relation

$$G(eV) = ta^2(eV)F(eV)$$

where the proportionality coefficient (the square of the EPI matrix element averaged over the Fermi surface) depends generally speaking on the energy. In many cases, however, this dependence is weak and it can be assumed that the principal maxima in the EPI MC spectra coincide in position with the singularities of the phonon states of the given metal.

Figure 1(b) shows the MC spectrum of a Cu-Be alloy containing 2.7 at.% Be, where, besides the EPI spectrum of the copper, a singularity is present in the region of 40 meV, i.e., at an energy higher than the maximum energy of the phonons in the copper, but lower than the energy of the first phonon maximum in beryllium. The energy position of this singularity agrees well with that of the local mode in the spectrum of the given alloy, a postion determined from experiments on inelastic neutron scattering.[8] This makes it possible to interpret the indicated singularity as the result of electron interaction with the local vibrations (LV) in the Cu-Be system.

Figure 2 shows the MC spectra for a number of samples with different Be contents. As expected, the LV peak increases in intensity and broadens with increasing beryllium concentration.

It is of interest to compare the intensity of the LV band, per Be impurity atom, with the integral intensity of the MC spectrum S_{spectr} of the EPI of Cu per matrix atom. S_{spectr} was determined from the experimental curves in the region of the phonon frequencies with the background subtracted by the usual procedure.[2,9] To separate the LV spectra, the background was linearly extrapolated from the energy region

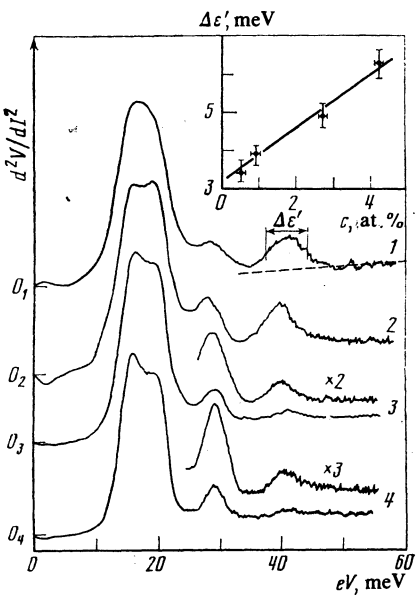

FIG. 2. Microcontact spectra of $CU_{1-c}Be_c$ alloys with different Be-impurity concentrations: 1) for $c = 0.042$, 2) 0.027, 3) 0.09, 4) 0.005. For spectra 3 and 4 are shown the additional spectra obtained by increasing the modulating voltage. Inset—concentration dependence of the width (at half-maximum) of the spectra of the local vibrations.

$eV > 60$ meV, where the contribution of the multiphonon processes is negligible.[1] In the calculation of the integral intensity S_{LA} of the LA singularity we used the same background extrapolation (see Fig. 1). This method of determining S_{LA} is, of course, approximate but is fully justified since the energy dependence of the background varies little in the region of the LA peak.[2] No significant errors are thus introduced by this procedure when S_{LV}/S_{LA} is calculated. We note that for each alloy the points on the concentration dependences of Figs. 1 and 2 are mean values of the corresponding parameter, obtained for several dozen curves corresponding to all the selection criteria (see, e.g., Ref. 9). The relative intensity of the LV band, determined in this manner is

$$c^{-1}S_{LV}/S_{spectr} = c^{-1}\int_{LV}\tilde{\alpha}^2(eV)F(eV)\,d(eV) \Big/ \int_{spectr}\tilde{\alpha}^2(eV) \times F(eV)\,d(eV) = c^{-1}\langle\tilde{\alpha}^2\rangle_{LV}N_{LV}\,(\langle\tilde{\alpha}^2\rangle_{spectr}N_{spectr})^{-1}. \quad (1)$$

Here N_{LV} and N_{spectr} are the numbers of the phonon states corresponding to the LV and the lattice excitations. The corresponding value of the relative intensity of the LV band, according to neutron-measurement data,[8] is

$$c^{-1}N_{LV}/N_{spectr} = c^{-1}\int_{LV}F(eV)\,d(eV) \Big/ \int_{spectr}F(eV)\,d(eV) = 3.6. \quad (2)$$

For microcontact spectra of alloys with Be concentrations 0.5, 0.9, 2.7, and 4.2 at.% the values of (1) are respectively 3.87 ± 0.2; 3.46 ± 0.15; $3.24^{-}0.4$ and 2.74 ± 0.15. The ratio $\langle\tilde{\alpha}\rangle_{LV}/\langle\tilde{\alpha}\rangle_{spectr}$ as $c \to 0$ is in this case close to unity, i.e., the average effectiveness of electron scattering by the vibrations of an isolated impurity center is approximately the same as by vibrations of the host lattice. With increasing concentration, the value of $c^{-1}S_{LV}/S_{spectr}$ decreases much more rapidly than would follow from the $f(c) = 1 - c$ dependence observed in Ref. 8 for (2). This decrease of the relative intensity with increasing impurity concentration is typical also of the high-frequency phonon peaks in the MC spectra of noble metals.[2,7] The mechanism of this phenomenon is not yet clear. Nonetheless, since a good correlation was observed for the MC spectra of the Cu-Be alloy between the intensity of the peak of the longitudinal phonons LA and LV peak, and the S_{LV}/S_{LA} plot shown in the inset of Fig. 1 is linear in the entire concentration region, it can be stated that the electrons are scattered by the LV and by the longitudinal acoustic phonons with approximately equal effectiveness that does not depend on the concrete structure or on the microcontact geometry.

The line shape in the LV and MC spectra is closer to Lorentzian (with broad wings) than to Gaussian. In addition, the LV band is as a rule asymmetric and smeared out on the high-energy side. Since, however, the shape of the function of the temperature and modulation smearing of the MC spectrum is close to Gaussian,[1] we calculated the true width $\Delta\epsilon^1_{LV}$ of the band (at half-maximum) by using the formula[2]

$$\Delta\varepsilon_{LV}'=[\Delta\varepsilon_{LV}^2-(5.44kT)^2-(1.72eV_1)^2]^{1/2}, \quad (3)$$

where Δ_{LV} is the observed width of the LV band, and V_1 is the effective value of the modulating voltage.

For the average line width of the LV in MC spectra, just as in the neutron experiments, we obtained a linear dependence of $\Delta\epsilon_{LV}^1$ on the concentration (see the inset of Fig. 2):

$$\langle\Delta\varepsilon_{LV}'\rangle=(3.1+72\ c)\ \text{MeV}. \quad (4)$$

The value of $\Delta\epsilon_{LV}^1$ obtained by us for an isolated impurity ($c=0$) is larger than follows from neutron experiments as $T,c\to 0$ (Ref. 10). The slope can also exceed the value obtained in Ref. 8 by 2.3 times, thus indicating that the impurities have a larger influences on the smearing of the LV in the MC spectrum.

The broadening of the LV in the MC spectra can be due to the influence of inhomogeneous mechanical stresses in the contact region. These stresses can reach the eleastic limit at the center of the contact and fall off smoothly towards the periphery. In addition, an effective "heating" of the phonon subsystem by an electron flux of high density is also possible. According to an estimate for the mode of nonequilibrium phonons fully trapped in the contact region,[11] their temperature can reach

$$kT_{\text{phonon}}\sim \text{eV}/4,$$

which yields $T_c\sim 110$ K for the LV of Be in a Cu matrix. This is still a relatively low temperature to broaden substantially the LV band as a result of anharmonicity.[10] This is also attested by the absence of a correlation between $\Delta\epsilon_{LV}$ and the background level outside the MC spectrum, which is proportional to the phonon heating in the contact region.[11]

We have investigated the energy region up to 120 meV for samples with Be concentration 2.7 at.%, with an aim of substantially broadening the LV band by anharmonicity. However, no singularities that are multiples of the LV energy were observed, accurate to 7–10% of the main LV peak in the MC spectrum. According to the theory,[12] singularities at double the phonon frequencies can be brought about by several mechanisms:

1. The main contribution to the two-phonon band should be made by processes of successive emission of two phonons by one and the same electron. The order of magnitude of this contribution is $(d/l_e)^2$, and relative to the main peak the value of d/l_e in our case is 4–9% of the intensity of the LV band.

2. Coherent simultaneous emission of two phonon is made possible by the small nonadiabaticity of the relative motion of the electron and phonon subsystems. The intensity of these processes is proportional to two small parameters: d/l_e and the adiabatic factor $eV/\epsilon_f\approx\hbar\omega_{LV}/\epsilon_f\ll 1$, and is usually much less than two-phonon process described in item 1.

3. Finally, either emission of two phonons by one electron, or else formation of a "biphonon" as a result of anharmonicity, is possible. The probability of these processes as $T\to 0$ contains besides the factor d/l_e, the de Boer parameter (the square of the ratio of the amplitude of the zero-point ovibrations to the lattice constant), and is therefore negligibly small.

Consequently, to observe singularities in the $2\hbar\omega_{LV}$ regions it is necessary to increase substantially the sensitivity of the MC-spectrum measurements at high energies. This sensitivity is limited mainly by the electric fluctuations generated by the microcontact.

We can thus conclude that MC spectroscopy allows us to investigate the interaction of electrons with LV due to light impurity atoms in a metallic lattice. It can be assumed that other phonon-spectrum perturbation induced by various types of lattice defects are also reflected in the microcontact functions of the EPI.

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