# Positron spectroscopy of nanoscale regions of homogeneous atomic and magnetic orderings in strongly correlated alloys


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**Actuality.** Today, the most promising materials of electronic, energy, and aerospace industries are developed not only taking into account quantum effects due to the nanometre level of the element base of devices, but also complex collective phenomena in the electrons' subsystem of solids and other types of lattice perturbations (phonons, magnons, etc.). These effects can significantly change the electrical, optical, magnetic and even mechanical properties of the system with relatively small external influences, and therefore their presence can be detected by changing many physical characteristics. However, the most of these changes are caused by the modification of the electronic structure, for example, due to strong electron-electron correlations. A very limited number of methods allows an experimental observation of these changes in the electronic structure. Such methods reproduce the Fermi surface in strong magnetic fields (based on Haas–van Alphen and Shubnikov–de Haas effects) or permit a direct observation of the band structure (using angle-resolved photoemission spectroscopy (ARPES)). Both of these methods require the magnetic fields (including very strong ones), low temperatures and, in the case of ARPES, ultra-high vacuum, and they are only applicable to systems with a well-defined band structure and Fermi surface. All these experimental complexities and natural limitations are absent in the method of positron spectroscopy, namely, in the method of angular correlation of electron positron annihilation radiation (ACAR).

**Models of ACAR.** This method involves partial hardware integration of the momentum distribution of electrons by energy and by components of the wave vector, which lies in the plane of the slit of the annihilation gamma rays' detector, which allows to analyse poorly defined (spread) momentum distributions of electrons, including the vicinity of the Fermi surface, for example, for nonstoichiometric alloys. The use of spin-polarized positrons additionally allows determining in magnetic materials separately for each of the directions of the electrons' spin the distribution of their by pulses. In addition, this method allows to determine the size and concentration of defects in crystalline or non-crystalline matter and to study their electronic properties. Thus, the theoretical consideration of the influence of some many-particle effects on the momentum distributions of conduction electrons recorded by the ACAR method (Figs. 1, 2) is actual (note, that contributions of core electrons, positronium and other paths of annihilation are not considered here).

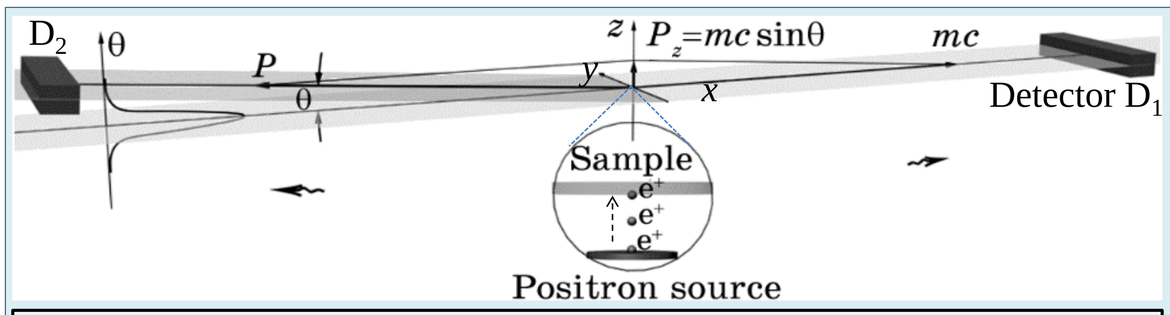


**Fig. 1. Sketch of the angular correlation spectrometer of annihilation radiation.**

**The band electrons contribution in the free electrons approximation** | Not considered contributions of localized electrons

$$I^{theor}(\theta) = I_P\left[\frac{\theta_F^2-\theta^2}{2} + A_B T ln\left(1+exp\left\{-\frac{\theta_F^2-\theta^2}{2A_BT}\right\}\right)\right] + \sum_{j=1}^{3}\frac{I_G^j}{\sigma_j\sqrt{2\pi}}exp\left\{-\frac{\theta^2}{2\sigma_j^2}\right\}$$

$I^{theor}(\theta)$ is the intensity register by ACAR, $\theta$ is the difference between the scattering angle of the annihilation $\gamma$-quanta and $2\pi$, $I_P$, $I_G^j$ are the amplitude coefficients of the parabolic and Gaussian parts of the spectra, respectively, $\sigma_j$ are the dispersions, $\theta_F$ is the angle corresponding to the Fermi momentum, $A_B$ is the renormalized Boltzmann constant, and $T$ is the temperature of the sample.

**The band electrons contribution in the Hubbard model for the substitution binary alloys:**

$$I_{be}(\theta) \propto f(\theta) = \frac{1}{A}\int_{P_z-\Delta P_z/2}^{P_z+\Delta P_z/2} dk_z \int_{-\Delta P_y/2}^{\Delta P_y/2} dk_y \int_{-\infty}^{+\infty} dk_x n_e(k_x,k_y,k_z)$$

$$n_e(\mathbf{k}) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} dE \sum_{s=1,2} f(E,T)\,\mathrm{Im}\left(\left\langle G_{es}^{A}(\mathbf{kk},E)\right\rangle\right)$$

$$\left\langle G_{es}^{A}(\mathbf{kq},E)\right\rangle = \frac{\delta_{\mathbf{kq}}}{E^- - E_s(\mathbf{k}) - \Sigma_s(E)};\ P_z \approx mc\theta$$

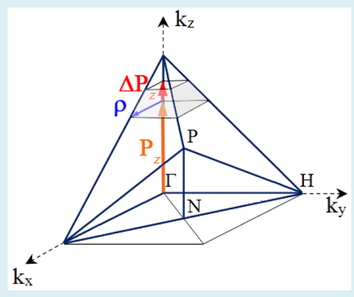


**Fig. 2. Part of the 1st Brillouin zone of the b.c.c. alloy for coordinates $\{k_x, k_y, k_z\}>0$ with integration layer.**

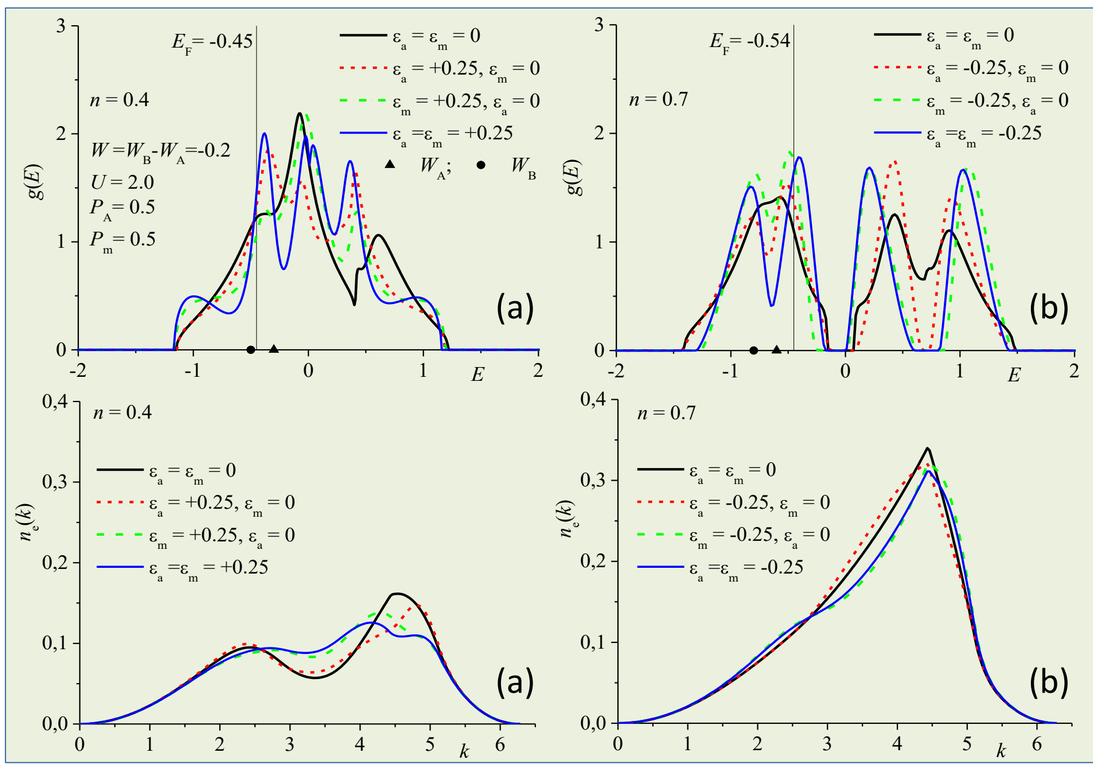


**Fig. 3. Densities of electronic states for b.c.c. alloys with different atomic and magnetic short-range orderings and two values of electron concentration *n*.**

**Fig. 4. Dependences of $n_e(k)$ (~$I_{be}(\theta)$ (ACAR)) from modulus of electron wave vector *k* for b.c.c. alloys with different atomic and magnetic short-range orderings and two values of electron concentration *n*.**

**Results of numerical calculations**. Energy is measured in units of half-width of the energy zone of a one-component crystal, localized magnetic moments are measured in Bohr magneton $\mu_B$. For the absence of a magnetic field, the probabilities of finding the projections of localized magnetic moments in the selected site (0) are the same: $P_0^{\mu+}=P_0^{\mu-}=0.5$. Equilibrium values of magnetic moments ($\mu^{\pm}$) and orders parameters ($\varepsilon_m$, $\varepsilon_a$) are found from the condition of free energy minimum. The parameters of short-range atomic ($\varepsilon_a$) and magnetic ($\varepsilon_m$) orders determine the sizes *d* of regions of corresponding orderings in the investigated strongly correlated alloys.

The sizes of regions with homogeneous magnetic order:

$$\frac{d}{a} = 2k = 2\cdot\begin{cases} \ln(1/2)(\ln P_{j\,0}^{\mu_{\lambda'}^-/\mu_\lambda^-})^{-1}, & \text{for } \varepsilon_m>0 \text{ (s-FM, FM)},\\ 1, & \text{for } \varepsilon_m\cong 0 \text{ (SG)},\\ \ln(1/2)(\ln P_{j\,0}^{\mu_{\lambda'}^+/\mu_\lambda^-})^{-1}, & \text{for } \varepsilon_m<0 \text{ (s-AFM, AFM)}, \end{cases}$$

$$P_{j\,0}^{m_{\lambda'}/m_\lambda} = P_j^{m_{\lambda'}} \pm \varepsilon_{j\,0}^{\mu_{\lambda'}^-\mu_\lambda^-}/P_0^{m_\lambda}\quad (\varepsilon_{j\,0}^{\mu_{\lambda'}^-\mu_\lambda^-}\equiv\varepsilon_m, \lambda=[A,B], m_\lambda=\mu_\lambda^{\pm}).$$

For atomic order one can replase $\mu_\lambda^+\to A$, $\mu_\lambda^-\to B$, $\varepsilon_m\to\varepsilon_a$).

At the framework of the single-band Hubbard model for substitution binary alloys $A_xB_{1-x}$ (in our case $x=P_0^A=0.5=1-x=P_0^B$) with strong electron correlations, the changes in momentum distributions of electrons are investigated depending on the presence and magnitude of pair correlations in the arrangement of atoms of different types and/or in the orientation of electron magnetic moments at nearest sites. These correlations determine electrons ground state, their kinetic and thermodynamic properties. As shown, at temperature of 0 K and in crude estimation $k\approx P_z$, the effects of pair correlations on the electrons momentum distributions ($n_e(k)$, Fig. 4) are clearly observed. However, they are smaller than ones on density of electronic states ($g(E)$, Fig. 3). The Figs. represent the effects of non-equilibrium atomic and magnetic short-range orders and their simultaneously act on mentioned electrons energy and momentum distributions in comparison with fully disordered alloys (black lines). The magnetic separation and order ($\varepsilon_m=\pm0.25$, i.e. ferromagnetic (FM)/antiferromagnetic (AFM) states) have more influence on these electrons distributions due to strong electron-electron correlations than corresponding ordering in atomic subsystem. With the averaged electron concentration *n* increasing, the effect of magnetic and atomic orderings on electrons momentum distributions $n_e(k)$ is decreased.

**Conclusions**. As shown for substitution binary alloys, the electrons momentum distribution $n_e(k)$ is sensitive to many-body effects, including atomic and/or magnetic short-range orderings and strong electron-electron correlations, which allows to use ACAR for mentioned effects experimental identification. Particularly, it is possible to recover from ACAR experimental data, corresponding to a band electrons contribution, the equilibrium values of correlation parameters ($\varepsilon_a$ and $\varepsilon_m$), and estimate not only the linear size of regions of homogeneous short-range atomic ordering, but the analogous characteristic of nanosize magnetic regions. As determined, the ACAR is more sensitive to magnetic ordering than to atomic one. The sensitivity of electrons momentum distributions to magnetic and atomic orderings is decreased with the averaged electron concentration increasing, and it is smaller than one for density of electronic states of strongly correlated alloys.