Anomalous low temperature diamagnetism in compounds and alloys

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Abstract

Applications of pronounced singularities of orbital magnetic susceptibility χ, related to the bands degeneracy points close to the Fermi level E_F, are demonstrated experimentally and theoretically. Such degeneracy points are found to be responsible for the anomalous diamagnetism in $CaPb_3$ and $YbPb_3$ compounds at low temperatures. In Al the anomalies in χ were realized by doping of Al with Li and Zn. The appearance and smearing of these anomalies with temperature, impurities, and quenched vacancies are analyzed, and corresponding band structure and scattering parameters have been determined.

Key words: Anomalous diamagnetism; bands crossing; $YbPb_3$; Al alloys

Within the general theory of orbital susceptibility χ of Bloch electrons [1], it was shown [2–4], that the χ can diverge when the Fermi level E_F passes through the point of the energy bands contact. These diamagnetic singularities in χ are expected to be very sensitive to the thermal excitations and the finite life-time of quasiparticles [2,3,5]. With accompanying theoretical analysis of amplitudes and locations of the features in χ, this provides avenue to reveal corresponding parameters of the conduction electrons scattering and fine details of the band structure $E(\mathbf{k})$. Here we are dealing with complex metallic systems exhibiting anomalous low temperature diamagnetism, specifically aluminium alloys with a strong disorder, and intermetallic compounds $CaPb_3$ and $YbPb_3$, having a substantial admixture of d- and f-states in the band structure.

In Al the degeneracy point in $E(\mathbf{k})$, which cause the diamagnetic singularity in χ, is located below E_F. At low temperatures, this singularity looks like a sharp deep in the background paramagnetic susceptibility of Al, which was observed [3,6] in alloys with Mg and Zn, corresponding to the mean valency $Z = 2.944$ (see Fig. 1). These solid solutions can be obtained just by quenching, and the anomalous diamagnetism is much dependent on heat treating of the samples.

In Al–Li alloys the bands degeneracy point is reached within the solubility region at room temperatures. We observed the deep in $\chi(x)$ at x(Li)=2.8 at.%, i.e. approximately at the same mean valency as in the Al–Mg and Al–Zn alloys (Fig. 1). But in contrast to them, in the Al–Li alloys the position of the deep is independent on heat treating, and these $\chi(Z)$ data can be used as the reference ones. Following Ref. [3] and evaluating amplitudes of the deep, we obtained the scattering parameter $d\Gamma/dx$(Li)=125±10 K/at%, which appeared in accordance with the relative effect of impurities on the resistivity of Al.

The effect of heat treating on χ in non-equilibrium Al–Zn alloys is demonstrated in Fig. 1 for the sample with x(Zn)=4.5 at.%, which was water quenched from the homogeneous state at 720 K and then annealed for 3 hours at ≃400 K. The degenerate band states are presumed to be nearly destroyed by the extended defects, such as precipitates and Guinier – Preston zones, which arise in this system during quenching. In Fig. 1 we

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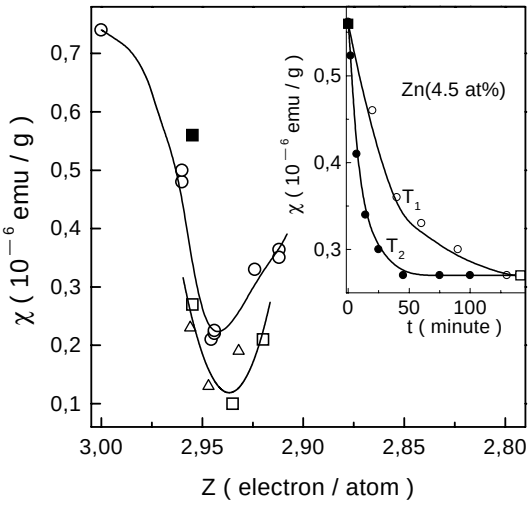

Fig. 1. Magnetic susceptibility of the Al-based alloys at T=78 K as function of mean valency (Al-Li (◯); Al-Zn: annealed (□), (△)[6], quenched (■)). In inset, the susceptibility of Al+Zn 4.5 at.% sample as function of the time of annealing at temperatures T_1=400 K (○) and T_2=440 K (●).

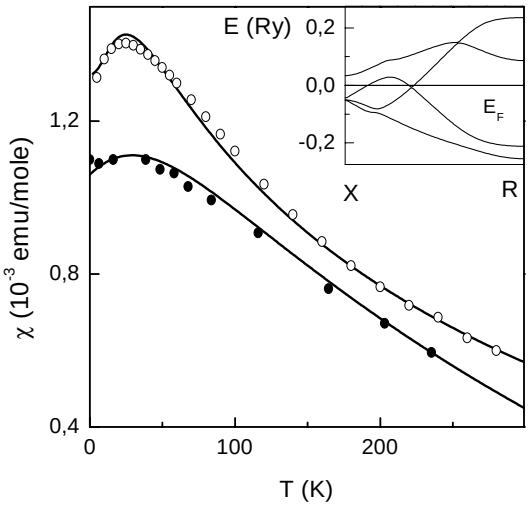

Fig. 2. Temperature dependence of the magnetic susceptibility of $CaPb_3$ (●, Ref. [7]) and $YbPb_3$ (○, present experiment). The solid lines correspond to the calculated χ. The $E(\mathbf{k})$ spectrum in the XR direction of the Brillouin zone is shown in the inset.

present kinetics of annealing of these defects, which is accompanied with regaining of the anomalous diamagnetism, probably due to elimination of non-equilibrium vacancies as a result of artificial aging. Comparing the time scales of the susceptibility changes $\chi(t)$ shown in Fig. 1 for different annealing temperatures, one can roughly estimate the activation energy of this process as $W \sim 0.5$ eV. The further phase decomposition in Al–Zn system is governed by substantially longer relaxation time.

Low temperature anomalies of χ are also observed in the isovalent intermetallic compounds $CaPb_3$ [7] and $YbPb_3$ (see Fig. 2). In the present work the measurements of $\chi(T)$ are carried out on the high quality single crystal of $YbPb_3$. In order to locate the corresponding degenerate band states, *ab initio* electronic structure calculations are performed for both compounds by using the fully-relativistic full-potential LMTO method [8]. In the calculations, the divalent state of Yb was assumed in $YbPb_3$, and bands degeneracy points were revealed in the close vicinity of E_F in both compounds (see the inset in Fig. 2). The calculated band parameters were used for a theoretical evaluation of the singular part of χ, according to Ref. [4]. In order to fit the theoretical χ to the experimental data, the *ab initio* calculated local positions of E_F (relative to the bands crossing energy) had to be only slightly adjusted over 1 mRy range, i.e. well within the accuracy limits for the *ab initio* calculations, and the values of E_F were determined as 1.1 mRy and 0.4 mRy for $CaPb_3$ and $YbPb_3$, respectively, at low temperatures. Also the scattering parameters Γ were found to be 50 K and 16 K (respectively for $CaPb_3$ and $YbPb_3$), which ensured the correct behaviour of the calculated $\chi(T)$ at low temperatures. As is seen in Fig. 2, the calculated $\chi(T)$ appeared to be in agreement with the experimental data in both $CaPb_3$ and $YbPb_3$ compounds.

In conclusion, we demonstrate that the singular diamagnetic contributions to χ, originating from the degenerate band states, can serve as the efficient tool for a quantitative analysis of the electronic structure itself, and also for monitoring of various processes and perturbations which can cause a reconstruction of $E(\mathbf{k})$.

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