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Positron Annihilation with Electrons of Admixture Atoms in Some Binary Nickel Alloys

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Weighted difference curves have been shortly presented as a proper tool allowing to analyze in another way two-detector Doppler spectra regarding positron annihilation with admixture electrons in some nickel alloys. The analysis of low and middle momentum parts of Doppler spectra for investigated alloys reflects the annihilation of positrons with admixture electrons. There is no distinct signal of annihilations with admixture electrons from the positron lifetime data for well-annealed samples.

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1. Introduction

On the 32nd Polish Seminar on Positron Annihilation we have presented the results of investigations of binary nickel alloys containing 1 at.% of Ge, Zn, In, Zr, Pb, Ti, and Sb by positron annihilation methods [1]. For cold-rolled samples before measurements there were some small but distinct differences between the Doppler spectra for different alloys. It has been suggested that the values of the defect component in the measured positron lifetimes could be connected with dimensions of admixture atoms as well as with vacancy-impurity binding energy which influence the number of such pairs in an alloy. The proposal has not been based on the Doppler measurements even though the differences in the Doppler distributions were observed. In the present study, we try to apply the Doppler spectra to confirm the presence of vacancy-impurity pairs. Additional series of

measurements for well-annealed alloys and their elements have been performed. Some measurements of Doppler spectra for cold-rolled samples were repeated and the newest results of measurements are now the object of analysis.

In the procedure of identification of vacancy–impurity pairs it is necessary to recognize atoms from which the electrons have taken part in the annihilation. Core electrons are the most suitable for that because they are only little affected by chemical environment. Their large energies are only negligibly disturbed by surrounding atoms and, which is more important, they are usually unique for each type of atom [2, 3]. High-momentum annihilation events, valuable for the identification, are rather not very numerous because of strong repelling forces between positrons and atom cores. Therefore, the success of applying procedure of identification of elements based on the measurement of high-momentum part of energy distribution of annihilation depends strongly on the quality of background selection and resolution of the measuring system. The peak to background (PB) ratio of our coincidence spectrometer equals one million, and the estimated energy resolution is 1.25 keV [4, 5].

2. Experimental

Investigated alloys were produced from 4N purity components by melting them in an induction furnace. Plates of 2 mm thickness were annealed under vacuum for three days at 1000°C. Composition and homogeneity of alloys as well as content of residual impurities were tested by laser and electron beam microanalyses. The samples were cold-rolled in steps of 0.1 mm to a thickness reduction of 50%. The well-annealed samples of elements (Ti, V, Cr, Fe, Ni, Cu) used in experiment were supplied by Jonson Matthey. They are Specpure class and it means that they are at least 4N. The remaining materials (Ge, Sb, Pb, In) were supplied by Goodfellow. They are of 5N purity class.

3. Results and discussion

The annihilation momentum distribution spans over nearly 6 orders of magnitude. This makes a detailed comparison of high-momentum region of spectra with a low number of counts difficult. A common way to overcome the problem is to use the ratio curves. However, the ratio curves are well suited to reveal differences only at medium and high momenta. Data belonging to high-momentum parts of spectra are usually strongly scattered due to low statistics. The most important differences between the momentum distributions for investigated alloys extend from 5 to 20 ($\times 10^{-3} m_0 c$). Therefore, we have used weighted difference curves ($\frac{N - N_{\text{ref}}}{\sqrt{N + N_{\text{ref}}}} = f(\frac{1}{2} p_L)$, where $N(\frac{1}{2} p_L)$ — the number of counts of the considered spectrum, $N_{\text{ref}}(\frac{1}{2} p_L)$ — the number of counts of the referenced spectrum, and p_L — the longitudinal component of annihilating pair in laboratory system) which

are more suitable in presenting the data at low and middle momentum [6]. In the weighted difference curves the difference of the number of counts of the considered curves is divided by the statistical error of counts of both curves ($\sqrt{N + N_{\text{ref}}}$). All the measured Doppler spectra were normalized to the equal number of counts within the range of 0 to $35 \times 10^{-3} m_0 c$.

The weighted difference between Doppler spectra for pure undefected admixture material (polycrystalline Zn) and matrix (Ni) is within the range of 0 to about 5 — positive and within the range of 5 to 20 ($\times 10^{-3} m_0 c$) — negative. The tendency of shifting of weighted difference curve for Ni(Zn) alloy from bulk Ni towards that characteristic of pure Zn can be observed in Fig. 1. Such tendency has been observed for all the investigated alloys, but its scale is different for different samples. The largest shift has been revealed in the case of Zr admixture.

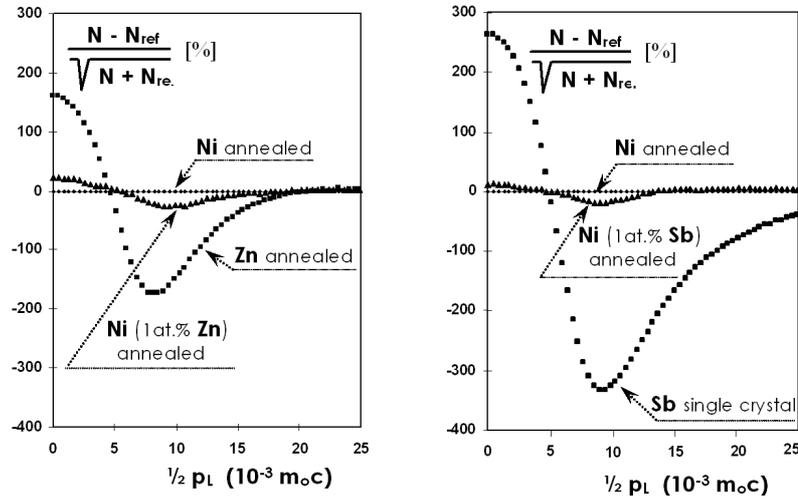


Fig. 1. Weighted difference curves for annealed Ni(Zn), Ni(Sb) alloys, Zn, and Sb.

Analysing lifetime data for well-annealed samples (Tables I and II) we can see that for all the samples the value of lifetime for the first component is near 108 ps, which is characteristic of pure nickel. The occurrence of the second lifetime component with intensity of a few percent can be explained by annihilations of positrons in air or some extra defects not annealed during the annealing procedure. The similar values of bulk lifetime ($\tau_B = \tau_1 \tau_2 / (\tau_1 I_2 + \tau_2 I_1)$) for well-annealed samples, close to the value characteristic of pure Ni, confirm validity of the used model.

Deformation (cold-rolling) of the samples is connected with generation of numerous defects. In the similar procedure of cold rolling of nickel samples Dlubek et al. achieved saturation of annihilation positrons trapped at vacancies for samples whose thickness has been reduced by 18% [7]. In our study the sample thickness has been reduced by 50%, therefore it is sound to assume that for deformed samples

TABLE I

The lifetime data for nickel alloys with 1 at.% of admixture.

Cold-rolled alloys					
Admixture	τ_1 (ps)	τ_2 (ps)	I_1 (%)	I_2 (%)	$\bar{\tau}$ (ps)
Zn	185.5 (2.6)*	299.3 (6.6)	71.7 (2.8)	27.1 (2.8)	234 (18)
Ge	173.8 (0.9)	317.4 (2.7)	73.3 (0.8)	25.6 (0.8)	227 (5)
Se	173.9 (1.6)	321.8 (4.9)	74.7 (1.3)	24.2 (1.3)	226 (10)
Zr	181.0 (1.0)	413.0 (5.0)	95.7 (1.0)	4.3 (1.0)	191 (7)
In	177.5 (1.0)	361.0 (13.9)	92.0 (0.8)	6.9 (0.8)	207 (7)
Sb	182.8 (1.1)	337.6 (7.8)	85.9 (1.1)	13.0 (1.1)	220 (8)
Pb	177.2 (1.3)	341.7 (9.0)	86.1 (1.0)	12.7 (1.0)	216 (9)

*Standard deviations are given in round brackets.

TABLE II

The lifetime data for nickel alloys with 1 at.% of admixture.

Annealed alloys			
Admixture	τ_1 (ps)	I_1 (%)	τ_{Bulk} (ps)
Zn	105.1 (0.5)*	100	105
Ge	108.8 (0.7)	99.2 (0.2)	110
Se	106.0 (0.5)	99.1 (0.2)	108
Zr	108.5 (0.6)	97.9 (0.2)	111
In	107.9 (0.5)	99.49 (0.2)	109
Sb	108.1 (0.5)	100	108
Pb	107.1 (0.4)	98.49 (0.1)	109

*Standard deviations are given in round brackets.

nearly all annihilations take place within vacancies. The lifetime measurements confirm this supposition. The average value of the first lifetime component for deformed samples equals 179 ps which corresponds well with the value of 180 ps of the average lifetime of positrons trapped in vacancies in pure nickel, calculated by Puska et al. [8]. The second component of considerably less intensity is characteristic of positrons trapped at defect complexes.

The tendency of shifting of weighted difference curves of alloys towards those characteristic of pure admixture, observed originally for cold-rolled samples, was observed for well-annealed samples too, but the scale of shift for deformed samples is much greater (Fig. 2). If the reduced probability of high-momentum annihilation, observed for all investigated cold-rolled alloys, is due to annihilations of positrons with admixture electrons, the rise in the number of annihilations with admixture electrons simply means the larger number of defect-admixture pairs.

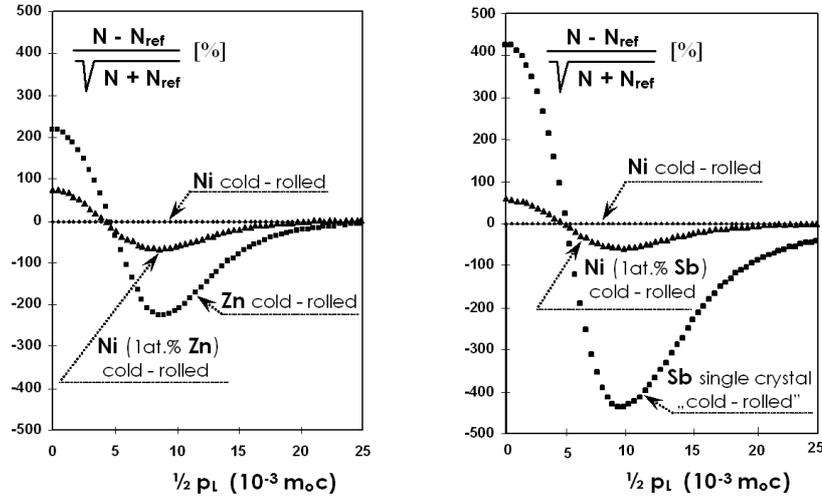


Fig. 2. Weighted difference curves for cold-rolled Ni(Zn), Ni(Sb) alloys, Zn, and Sb.

The crystal structure of samples, their defect state and purity, also influence the probability of high-momentum annihilations. The addition of only 1 at.% of admixture does not change the structure of Ni matrix, but modifies a little positions of matrix atoms in the nearest surrounding of admixture atom. This dimensional disproportion, as well as usually a different character of binding of admixture atoms in comparison with atoms of Ni matrix, favors the occurrence of vacancy–impurity pairs in such places, e.g. [9]. The large shifts of weighted difference curves of alloys towards those characteristic of pure admixture, observed for deformed samples, can be connected with an amount of the vacancy–impurity pairs. The shift could also be caused by defects in an alloy. The dominating character of the first lifetime component close to that characteristic of pure Ni reduces such eventuality for well-annealed samples into the background. The purity of samples can influence the Doppler distributions as well. In the investigation of alloys, with only 1 at.% of admixture, the 4N purity of alloy components do not guarantee independence of impurities indeed, but the observation of annihilations with admixture electrons is possible by analyzing the shifts of alloy curves from bulk Ni towards that characteristic of pure admixture.

It is worth to say that the results obtained by the two-detector Doppler broadening technique, presented in the form of weighted difference curves, reflects the presence of acts of annihilation with admixture electrons also for well-annealed samples. Such information cannot be gained from positron lifetime measurements. It means that for solving some problems of application of positron spectrometry in studies of the condensed matter structure, the Doppler coincidence method can be more useful than positron lifetime measurements.

4. Conclusion

The very promising possibilities of application of the two-detector Doppler broadening technique for studying the vacancy–impurity atom complexes in metallic systems have been demonstrated experimentally. The shifts of alloy weighted difference curves from bulk Ni towards that characteristic of pure admixture within the range of $(5–20) \times 10^{-3}m_0c$ are interpreted as a signal of annihilation of positrons with admixture electrons. There is no distinct signal of annihilations with admixture electrons from the positron lifetime data for well-annealed samples.

References

- [1] A. Baranowski, M. Kostrzewa, M. Szuszkiewicz, *Acta Phys. Pol. A* **99**, 329 (2001).
- [2] W. Deng, D. Pliszka, R.S. Brusa, G.P. Karwasz, A. Zecca, *Acta Phys. Pol. A* **101**, 875 (2002).
- [3] V.J. Ghosh, M. Alatalo, P. Asoka-Kumar, B. Nielsen, K.G. Lynn, A.C. Kruseman, P.E. Mijnarends, *Phys. Rev. B* **61**, 10092 (2000).
- [4] A. Baranowski, J. Beliczynski, M. Kostrzewa, M. Szuszkiewicz, *Nucl. Instrum. Methods A* **526**, 420 (2004).
- [5] M. Kostrzewa, Ph.D. thesis, Wrocław University, Wrocław 2004.
- [6] J. Gebauer, R. Krause-Rehberg, S. Eichler, F. Borner, *Appl. Surf. Sci.* **149**, 110 (1999).
- [7] G. Dlubek, R. Krause, O. Brummer, Z. Michno, T. Gorecki, *J. Phys. F, Met. Phys.* **17**, 1333 (1987).
- [8] M.J. Puska, R.M. Nieminen, *J. Phys. F, Met. Phys.* **13**, 333 (1983).
- [9] M.J. Ponnambalam, P. Jena, *Hyperfine Interact.* **20**, 65 (1984).