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COMPARISON OF THE ELECTRON MOMENTUM DISTRIBUTION OF SOLID AND LIQUID NI AS MEASURED BY POSITRONS

M. J. Fluss, L. C. Smedskjaer, and M. K. Chason

Materials Science Division, Argonne National Laboratory, 9700 South Cass Ave
Argonne, Illinois, USA. 60439.

Résumé.- Dans ce travail on compare les densités de moment électroniques mesurées par positrons qui s'éliminent dans un réseau parfait de Ni, dans les lacunes et dans le Ni *liquide. Les techniques expérimentales et les méthodes d'analyse* sont discutées.

Abstract.- This paper compares the measured electron momentum densities seen by positrons annihilating in the defect-free Ni lattice, in vacancies and in liquid Ni. The experimental techniques and analysis method are also described.

1. INTRODUCTION.- The use of positron annihilation spectroscopy has seen a rapid increase in its application to the study of metals. The gamma rays emitted by an annihilating positron-electron pair provide information about the momentum and density of the electrons in the metal. When an energetic positron (E>kT) enters a crystalline material it thermalizes rapidly and then propagates as a Bloch wave. The positron in the Bloch wave state, because of its delocalization, is very sensitive to small concentrations of lattice defects such as atomic vacancies or other similar open regions. Vacancies trap the positron in a localized state with a binding energy of ~1-3 eV. An important feature of this trapping process is that the signal provided by a positron-electron pair annihilating from the vacancy state is noticeably different from the signal produced by annihilations from the Bloch state. Sensitivity to this trapping is usually achieved at vacancy concentrations $>10^{-6}$. One might expect that in a vacancy the lack of a nucleus and its associated core (high-momentum) electrons would result in a narrowing of the momentum distribution of electrons sampled by the positron. This is, in fact, exactly the case: Positron-electron pairs annihilating from the Bloch state reveal a broader momentum distribution than do those annihilating from the vacancy-trapped state.

2. POSITRONS IN LIQUID METALS.- The understanding about positrons annihilating in liquid metals is not as complete as in the case of solid metals. First, the nature of the positron in the liquid must be established. For example, the question may be raised as to whether the positron is in a localized or delocalized state. If, as one might anticipate, the positron is trapped in the open regions of the liquid, then how does the local electron structure of this trapped positron compare with that of a positron trapped in a vacancy? A second, and very basic, question which will have to be addressed is whether the positron perturbs its environment and thus stabilizes the hole in which it becomes trapped. An answer to the latter question will most likely come from advances in theory; however, experimental measurements that examine the temperature dependence of the annihilation parameters of positron-electron pairs in the liquid state should help to resolve this problem. Indeed, in the case of solids this was the historical evolution of the current theoretical understanding of the temperature dependence of positrons annihilating from the Bloch state.

The present report focuses on the state of the positron in solid and liquid Ni. Additionally, some indications of the nature of the temperature dependence of positron annihilation in Ni will be

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3. SAMPLE CONFIGURATION.- Polycrystalline Ni disks (99.995 nominal wt.%) were ion implanted with 10^{14} atoms of ^{58}Co, a positron emitting isotope. The disks were contained in an Al_2O_3 crucible. The crucible was in turn supported on a double bore Al_2O_3 tube at the supporting end of which was located a Pt/Pt-10% Rh thermocouple. A resistance heated furnace with heat shields and wires constructed from Ta surrounded the sample. This all in turn was mounted in an ultrahigh vacuum ($\sim 10^{-9}$Torr) system. A more detailed description of the experimental procedures will be found in Fluss et al. /1/.

4. SOLID Ni DATA.- Positron experiments were performed on the solid sample before and after melting to serve as a control with regard to possible contamination problems. The sigmoidal curve in Figure 1 indicates the type of data obtained in these base experiments. At each temperature the 511 keV radiation energy spectrum of the annihilation gamma rays was measured using a Ge(Li) detector. These spectra all show a Doppler-broadening due to the momentum distribution of the annihilating positron-electron pairs.

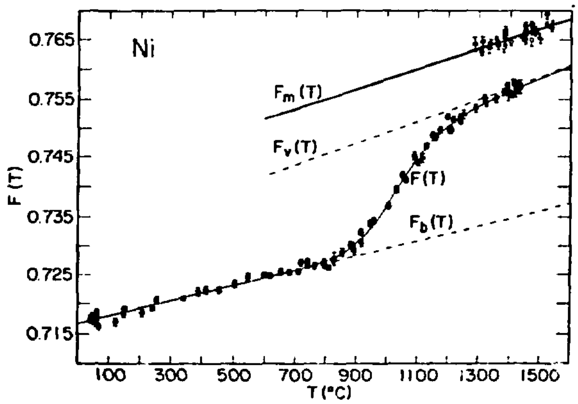

Fig. 1 : The positron annihilation Doppler broadening parameter F for solid and liquid Ni

The parameter F plotted in Figure 1 is a shape parameter derived from the Doppler-broadened spectra obtained at each temperature. Increasing F is associated with a narrowing of the momentum distribution of the sampled electrons. The lines $F_b(T)$ and $F_v(T)$ are the temperature-dependent shape parameters for positrons annihilating from the Bloch and vacancy-trapped states respectively in solid Ni. The transition between these two states is simply due to the exponentially increasing concentration of thermally generated vacancies. At the higher temperatures the sigmoidal curve approaches the $F_v(T)$ line as a result of the large concentration of vacancies, showing that almost all positrons annihilate while in the trapped state. The sigmoidal curve F(T) is derived from a model fit which takes the temperature dependences of these various states into account. The vacancy formation enthalpy for Ni has been determined from data such as these by Smedskjaer et al. /2/.

5. LIQUID Ni DATA.- When the Ni melts, the parameter F changes, indicating a narrower momentum distribution. Since liquid Ni could be easily supercooled, experiments were performed both above and below the melting point (1453°C). These data, also are shown in Figure 1. The data can be fit to a straight line, $F_m(T)$. It is interesting to note that the slopes of $F_m(T)$ and $F_v(T)$, are similar. The potential for providing useful temperature-dependence information is inherent in these data; however, more physically based parameters should be used. Moreover, the F parameter still contains the broadening associated with the instrumental resolution function, which is of the same order as the physical information itself.

6. DISCUSSION.- The Doppler-broadened spectra obtained from the three positron states (Bloch, vacancy-trapped, and liquid-trapped(?) states) provide information about the electron momentum densities. If the positron is delocalized (e.g., Bloch state) the electron momentum density obtained is that of the general lattice, while a localized positron yields information about the electron momentum density characteristic of the immediate

surroundings. Thus, a comparison between the vacancy-trapped and the Bloch states should show the differences between the electron momentum densities near the vacancy and that of the perfect lattice, while a comparison between the vacancy and the liquid will show first, whether the positron is *localized* or *not*, and second, (if the positron is indeed localized) the similarities between the sampled electron momenta around the vacancy and the "hole" in the liquid. A comparison of the three states of the positron described above in terms of the deconvoluted momentum distribution of electrons will be presented below. For the purpose of this example an isothermal comparison will be made at 1430°C, just below the melting point. Because almost all the positrons in the solid at this temperature are annihilating from the vacancy-trapped state, the temperature-dependent properties of the Bloch state have been extrapolated to this temperature.

The Doppler-broadened gamma ray spectra can be represented in terms of a resolution function (which was measured at 497 keV simultaneously with the 511 keV line) convoluted with a two-component physical spectrum consisting of a broad Gaussian component and a narrow parabolic component. The parabolic component represents completely delocalized electrons in the lattice, while the Gaussian component represents electrons more strongly localized near the nuclei. Both the widths and intensities of these components were left free to vary to obtain the best fits to the experimental data. The instrumental resolution is thus removed from the data. In Figure 2 we have plotted $p^2\rho(p)$, where p is the momentum and $\rho(p)$ is the momentum density as a function of the Doppler-shift energy in momentum space. The curves, which are derived from the experimental data, are normalized to unit area. Anisotropy cannot be considered owing to the polycrystalline nature of the samples.

Figure 2 shows the deduced positron-electron momentum distributions associated with the three states of the positron; the Bloch, vacancy-trapped, and liquid states, all at 1430°C. For comparison the momentum distribution measured for the electrons associated with the positron Bloch state at ~25°C is plotted as well. Certain similarities and differences can be described for the curves in Figure 2 in terms of the intensities and shapes of the deconvoluted components. The liquid and vacancy-trapped states differ from one another mainly in the intensity of the parabolic component (32% and 27% respectively) while the corresponding shapes (widths) of the parabolic and Gaussian components for these two states are nearly identical.

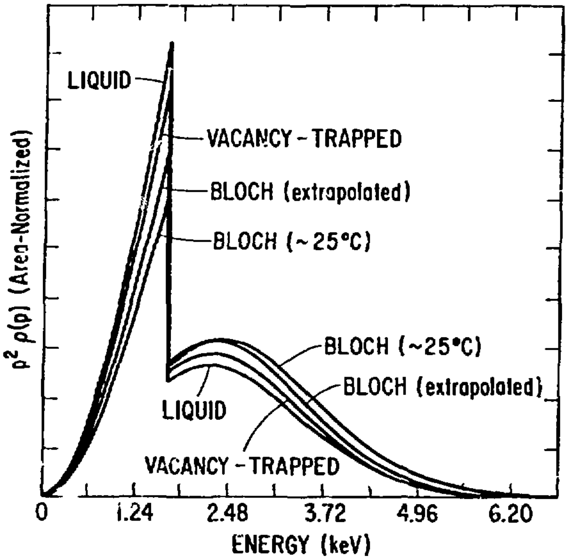

Fig. 2 : The electron momentum density function, $p^2\rho(p)$, as measured by the positron in the Bloch, vacancy-trapped, and liquid states, all normalized to unit area.

The extrapolated Bloch state has a Gaussian component whose width is broader than that of the vacancy-trapped (or liquid) state. In addition, the parabolic intensity is also significantly reduced to 20% for the Bloch state. The unextrapolated Bloch state (~25°C), however, has an even broader Gaussian width and an even smaller parabolic intensity. Since the liquid state differs from the localized vacancy state only by the relative intensities of the parabolic and Gaussian components, while the properties (widths) of the components which describe the two electron momentum groups are the same, one may conclude that the present description supports the idea of a localized state for the positron in liquid Ni.

The edge, representing the cutoff point of the parabola, in Figure 2 is found at p/mc=6.3 mrad, which is in reasonable agreement with calculations for the perfect Ni lattice /3-4/. It should be noted that Figure 2 is not a precise account of the directionally-averaged electron momentum density in Ni. It serves, however, as a means of comparison among the three momentum distributions of electrons shown here. The sharpness of the edges found in Figure 2 are partially a consequence of the model description of the Doppler-broadening spectrum. Indeed, to observe the details implied by Figure 2 it would be necessary to utilize instrumentation with significantly greater momentum resolution.

Recent experiments reported by Itoh and Suzuki /5/ on liquid Na and Ga, where positron angular correlation and Compton scattering experiments were combined, were interpreted as indicating positron trapping or localization in Ga, but not in Na. Lifetime experiments by Brandt and Waung /6/ suggested, however, that the positron was trapped both in liquid Ga and liquid Na. Analysis by direct comparisons of states, as presented here, may help to resolve this problem in the future.

In conclusion, a direct comparison of the as measured momentum distributions of annihilating positron-electron pairs points to a localized state of the positron in liquid Ni near the melting point. However, a significant question can be raised as to the degree of perturbation introduced by the very presence of the positron in the host liquid.

More detailed temperature-dependent deconvolution analyses have been undertaken. The goal is to answer some of the basic questions raised by the current work, and to determine the nature and value of the electronic structure information which the annihilating positron-electron pair is providing about the liquid metal.

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