[1]A. H. Bobeck, E. G. Spencer, L. G. Van Uitert, S. C. Abrahams, R. L. Barns, W. H. Grodkiewicz, R. C. Sherwood, P. H. Schmidt, D. H. Smith, and E. M. Walters, Appl. Phys. Lett. 17, 131 (1970).

[2]R. C. Le Craw, R. Wolfe, A. H. Bobeck, R. D. Pierce, and L. G. Van Uitert, J. Appl. Phys. 42, 1641 (1971).

[3]A. Rosencwaig, W. J. Tabor, F. B. Hagedorn, and L. G. Van Uitert, Phys. Rev. Lett. 26, 775 (1971).

[4]A. Rosencwaig and W. J. Tabor, J. Appl. Phys. 42, 1643 (1971); A. Rosencwaig, W. J. Tabor, and R. D. Pierce, Phys. Rev. Lett. 26, 779 (1971).

[5]H. Callen, Appl. Phys. Lett. 18, 311 (1971).

[6]S. Geschwind and J. P. Remeika, Phys. Rev. 122, 757 (1961).

[7]T. I. Barry, P. McNamara, and W. J. Moore, J. Chem. Phys. 42, 2599 (1965).

[8]L. G. Van Uitert, W. H. Grodkiewicz, and E. F. Dearborn, J. Amer. Ceram. Soc. 48, 105 (1965).

[9]See, for instance, J. F. Dillon and L. R. Walker, Phys. Rev. 124, 1401 (1961).

[10]W. P. Wolf, M. Ball, M. T. Hutchings, M. J. M. Leask, and A. F. G. Wyatt, J. Phys. Soc. Jap., Suppl. 17, B-I, 443.

Variation of Characteristic Energy Losses in the Curie-Temperature Region of Ni (111)

B. Heimann and J. Hölzl

Institut für Experimentalphysik der Technische Universität Clausthal, Clausthal-Zellerfeld, Germany

(Received 31 March 1971)

We have studied the temperature dependence of characteristic energy losses of slow electrons scattered from a Ni (111) surface. The volume loss energy shows an anomalous variation in the region of Curie temperature superimposed on a linear decrease from $T = 100$ to 700°C.

In an earlier publication[1] we studied the temperature dependence of secondary electron emission of polycrystalline Ni in the region of the Curie temperature T_C. In this paper we present the results of an experimental investigation of the spectrum of characteristic energy losses (CEL). In particular, CEL measurements will be discussed both as a function of temperature T ($100 \leq T \leq 700$°C) and primary energy E_p ($150 \leq E_p \leq 600$ eV).

In earlier studies[2,3] of the CEL spectra of electrons scattered from Ni, no variation in the energy of the CEL as a function of temperature was observed. Jordan,[3] in particular, reports CEL at 8.5 and 19.0 eV using a Ni (100) surface, which he interprets in accordance with Robins and Swan[4] as the surface and volume plasma energy losses, respectively. An additional loss at 28.0 eV is tentatively ascribed by Jordan[3] to both the combined bulk and surface plasmon exitation and an interband transition.

In the present experiment a Ni (111) surface is studied in a retarding-field analyzer with an energy resolution of $\Delta E/E = 0.5\%$. Special attention was given to the cleanliness of the crystal surface. The starting material was a 99.999% pure single crystal of Ni. Cleaning was done by oxidation, reduction, and thermal treatment.[5] Auger measurements and CEL, both being very sensitive to small surface contaminations, were used to demonstrate the cleanliness of the surface. All measurements were done in an ultrahigh vacuum system at pressures $p < 1 \times 10^{-10}$ Torr.

A typical energy spectrum, measured at $T = 200$°C and with a primary energy $E_p = 150$ eV is shown in Fig. 1. Some of the CEL detected by our experiments are clearly visible in this spectrum. Using various primary energies and the second derivative of the retarding-field curves, the following characteristic energy losses were observed at $T = 200$°C: $E_1 = 1.9$ eV; $E_2 = 3.4$ eV; $E_3 = 8.1$ eV; $E_4 = 11.0$ eV; $E_5 = 16.2$ eV; $E_6 = 19.1$ eV; $E_7 = 26.7$ eV; $E_8 = 33.2$ eV; $E_9 = 42.5$ eV; and $E_{10} = 67.0$ eV. The maximum experimental error of all loss energies measured was ±0.2 eV.

Following Jordan's interpretation, which is supported by his own optical measurements, it appears reasonable to attribute the energy losses $E_3 = 8.1$ eV and $E_6 = 19.1$ eV to the surface and volume plasma losses, respectively. With regard to the positions and intensities we attribute the loss energies $E_5 = 16.2$ eV and $E_9 = 42.5$ eV to the twofold surface and volume plasma losses, respectively. In this note we do not attempt to give an interpretation of the loss energies E_1, E_2, E_4, E_7, E_8, and E_{10}. The positions of these losses showed no variation with temperature.

The temperature dependence of the loss energies E_3 and E_6 as obtained with $E_p = 150$ eV is presented in Fig. 2. The following characteris-

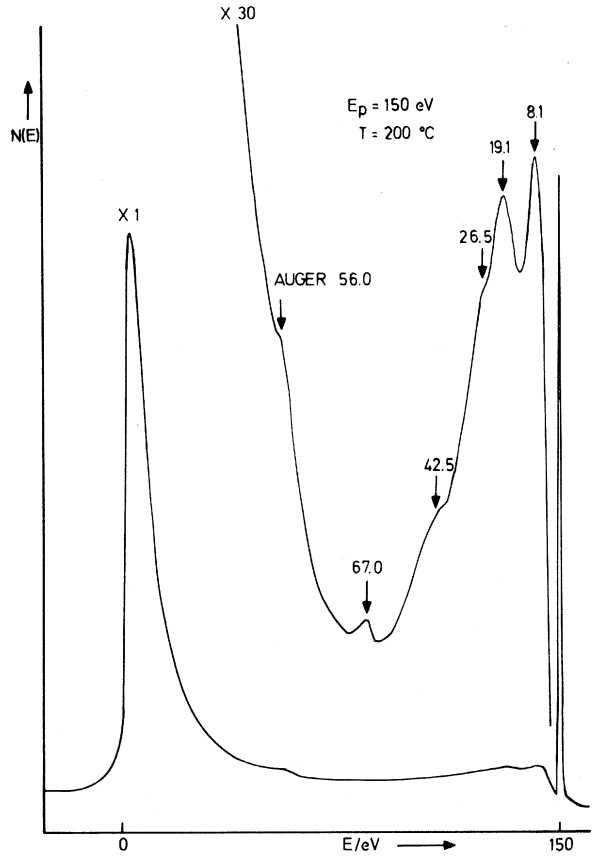

FIG. 1. Total energy spectrum of secondary electrons and spectrum of characteristic energy losses.

tics can be deduced from it: (1) There is a linear variation with temperature of the volume loss E_6 for temperatures $T \lesssim 280°C$ [temperature coefficient $\Delta E(T)/\Delta T = 2.9 \times 10^{-3}$ eV/°C]. (2) In the region of the Curie temperature T_C there is a marked deviation from this linearity. (3) Within the error of this measurement (±0.1 eV) the value of the surface energy loss E_3 is constant.

The following observations are important for the interpretation of the temperature variation of the energy E_6: (1) A closer examination of the loss spectra reveals, on the low energy side of the peak $E_6 = 19.1$ eV, the twofold surface loss $E_5 = 16.2$ eV which is clearly detectable at a primary energy $E_p = 150$ eV. With increasing temperature the intensity of E_6 decreases strongly but not the intensities of E_3 and E_5. This fact has been confirmed by numerous measurements. It causes an apparent shift of loss energy E_6 with temperature towards lower energies. (2) The intensity of peak E_5 decreases strongly with E_p so that E_5 is unnoticable above $E_p = 400$ eV. (3) $\Delta E(T)/\Delta T$ of E_6 is essentially constant (1.7×10^{-3} eV/°C) above $E_p = 400$ eV after a decrease between $E_p = 150$ eV and $E_p = 400$ eV.

At present it cannot be excluded with certainity that the decrease in the position of the energy loss E_6 is caused by temperature-dependent differences of the intensities of superimposed peaks. Quantitatively, however, it is difficult to reconcile such an interpretation with the observations (2) and (3). Therefore the temperature coefficient as observed at high primary energies may be attributed to a true temperature dependence of the loss energy E_6. This dependence is considerably larger than that due to the decrease of the electron density with temperature.

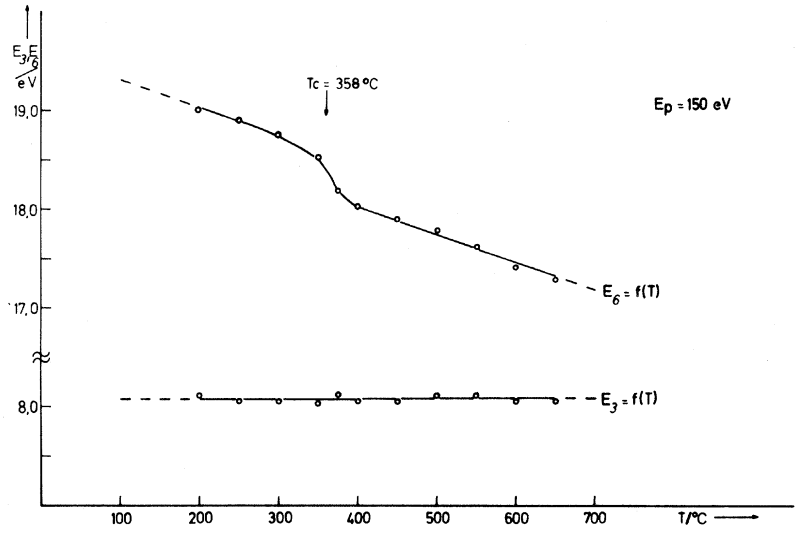

FIG. 2. Temperature dependence of surface loss E_3 and volume loss E_6.

Further evidence for a true temperature shift $\Delta E(T)$ of the volume loss energy is the observation of a temperature shift $2\Delta E(T)$ of the loss energy E_9 which is believed to be the twofold volume energy loss.

The nonlinear variation of the energy position of E_6 with temperature in the region of Curie temperature T_C ($280 \lesssim T \lesssim 400$°C) shows that the magnetization state of a crystal influences the CEL significantly. This influence may be understood as follows: It is widely accepted[3] that $4s$ and $3d$ electrons contribute to the volume plasma loss of Ni. The plasma energy calculated on the basis of a collective free plasma motion of one $4s$ and nine $3d$ electrons per atom is 35.4 eV. This value is depressed to the observed value of 19.1 eV through a strong coupling between highly energetic single excitations of $3d$ electrons and the plasmon fields.[3]

In the ferromagnetic state of the crystal an exchange splitting ($\Delta E_d \approx 0.4$ eV) of the $3d$ band has been predicted[6] and observed.[7,8] This splitting vanishes above T_C[6] associated with changes of the energy bands near the Fermi level such that the density of states reaches a sharp maximum corresponding to $3d$ band states immediately at the Fermi level.[6,9] This causes an increase of the excitation probabilities of single electron excitations from levels near the Fermi energy. Considering the above remarks a shift of the volume loss towards lower energies is to be expected.

There are other explanations of the anomalous variation of loss energy E_6 in the region of T_C such as a direct influence of the state of magnetization on the energy of the volume plasmon excitation. The interpretation presented here, however, appears to be the most reasonable one.

As to the striking temperature independence of surface-loss energy E_3 in contrast to the clear temperature dependence of the volume-loss energy, no preferred explanation can be given at present.

In addition to the CEL spectra we have measured the peak of true secondary electrons at 3.2 ± 0.1 eV and its temperature variation. Within the experimental error the energy position of this peak was found to be constant in the temperature range $100 \leq T \leq 700$°C. This demonstrates that the presence or absence of magnetic stray microfields near the surface of the crystal cannot alter the energy position of the loss peaks measured in our experiment.

We have benefitted greatly from stimulating discussions with E. Bauer. Support by the Deutsche Forschungsgemeinschaft is gratefully acknowledged.

[1]J. Hölzl and B. Heimann, Phys. Status Solidi 24, 651 (1967).

[2]J. B. Borowskii, A. N. Kabanov, Yu. M. Kushnir, and V. V. Schmidt, Zh. Eksp. Teor. Fiz. 38, 1383 (1960) [Sov. Phys. JETP 11, 997 (1960)].

[3]L. K. Jordan, thesis, Georgia Institute of Technology, 1969 (unpublished).

[4]J. L. Robins and J. B. Swan, Proc. Phys. Soc., London 76, 857 (1960).

[5]L. H. Germer and A. E. Mac Rae, J. Chem. Phys. 37, 1382 (1962).

[6]E. J. Zornberg, Phys. Rev. B 1, 244 (1970).

[7]M. Shiga and G. P. Pells, J. Phys. C: Proc. Phys. Soc., London 2, 1847 (1969).

[8]C. G. Windsor, R. D. Lowde, and G. Allan, Phys. Rev. Lett. 22, 849, 1162(E) (1969).

[9]J. Callaway and H. M. Zhang, Phys. Rev. B 1, 305 (1970).