Change of α-Particle Stopping Power in Ni at the Curie Temperature*

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(Received March 3, 2003)

Experiments showing that the ion–solid state interaction characteristics change at T_C, the Curie temperature, were recapitulated. It was shown that the experimental results for the increase in the α-particles stopping power in Fe and Gd foils can be approximately described by the Bethe theory of stopping power with Stoner model for band ferromagnetism. The experimental result for increase in 5.486 MeV α-particles stopping power in 0.89 mg/cm^2 Ni foil after transition from ferro- to paramagnetic phase at T_C was presented and it was shown to be in a good correlation with the previous data and with the theoretical evaluation.

PACS numbers: 34.50.Bw, 61.85.+p

1. Introduction

It is commonly agreed that at normal conditions some basic characteristics of ion–solid interactions like stopping power are independent of temperature in semiconductor and metallic targets, both amorphous and crystalline [1–3]. Within the linear response theory the temperature dependence of the stopping power goes entirely through the Fermi–Dirac momentum distribution of the occupation probability which enters the dielectric response function for the medium. It was shown [4, 5] that the energy loss can be effectively temperature dependent only at very high temperature of 10^6-10^7 K.

Other standard characteristics of ion–solid interaction like experimental energy loss straggling and theoretical nuclear encounter probability (NEP) are dependent on temperature; straggling is proportional to T [6] and NEP is based on

*supported by Uniwersytet Łódzki, grant 505/272 (2003)
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the Debye–Waller thermal factor. The angular Rutherford backscattering (RBS) spectra ($\chi_{\min}$ — minimum yield and $\psi_{1/2}$ — width at half minimum) and energy RBS spectra (χ_{min}) and also the surface peak size increase with temperature.

Moreover, it was shown that the structural, electric, and magnetic phase transitions can modify the characteristics of ion–solid interaction.

Frenkel et al. [7] showed that the transition through the melting temperature modifies RBS spectra allowing for analysis of surface melting. Lagare and Umakantha [8] found an anomalous 3% increase in the energy loss for 942 keV electrons in $BaTiO_3$ (ferroelectrics) at the Curie temperature of $T_C = 400$ K. The increase was explained by vanishing ferroelectricity at the Curie point. Recently Nickel et al. [9, 10] showed that the energy loss of 5 to 6 MeV alpha particles increases by 1.5% in 5.47 mg/cm^2 Fe ($T_C = 1043$ K) and by 0.2% in 9.53 mg/cm^2 Gd ($T_C = 297$ K) target after phase transition from ferromagnetic state below T_C to paramagnetic state above T_C. In an unpublished paper Nickel et al. [11] reported on 3.5% increase in He-like charge state fraction in charge distribution of 250 MeV/amu Bi ions transmitted through 1 g/cm^2 polycrystalline Gd when passing the Curie temperature T_C. Also, they found that above 21°C the energy loss increases in equidistant steps at temperatures corresponding to decrease in the Gd magnetisation value by a factor 2.

In this work we want to concentrate on the questions of how does the electronic stopping power of He ions in Ni foils change when the foil temperature increases and passes $T_C = 631$ K — the Curie temperature for transition from ferro- to paramagnetic phase?

2. Theoretical discussion

Within the Bethe theory the *random* stopping power is proportional to the number of electrons in the given shell $n_i \approx Z_a f_{i0}$ (f_{i0} is the dipole oscillator strength) and to the Bethe logarithm as

$$S_e \propto Z_a \ln \frac{E_m}{I} \approx \sum_i n_i \ln \frac{E_m}{\hbar\omega_i}, \tag{1}$$

where $\hbar\omega_i$ is the i-th shell binding energy, $E_m = 2mv^2 = 4Em/M$ is the maximum energy transferred in a collision from ion of mass M and an energy E to an electron of mass m. v is the ion velocity, Z_a is the target atom atomic number and I is the Bethe mean excitation energy defined as

$$\ln I = \sum_i f_{i0} \ln \hbar\omega_{i0}. \tag{2}$$

From Eq. (1) the difference Δ between the stopping power in a ferromagnetic $S_e(f)$ and paramagnetic $S_e(p)$ phase, related to $S_e(p)$ is given by

$$\gamma = \frac{\Delta}{S_e(p)} = \ln \frac{E_m}{I^f} / \ln \frac{E_m}{I^p} - 1. \tag{3}$$

The decrease in 5.5 MeV α-particle relative stopping power γ as measured by Nickel [9] ($\gamma = -1.5\%$ for Fe and $\gamma = -0.2\%$ for Gd) can be interpreted within the Bethe theory as due to increase in the mean excitation energy of the target atoms from I^{p} in paramagnetic phase to $I^{\mathrm{f}} = \alpha I^{\mathrm{p}}$ in ferromagnetic phase. $I^{\mathrm{p}}_{\mathrm{Fe}} = I_{\mathrm{Fe}} = 286$ eV and $I^{\mathrm{p}}_{\mathrm{Gd}} = I_{\mathrm{Gd}} = 591$ eV. Within the Bethe theory in the ferromagnetic state the mean excitation energy should be $I^{\mathrm{f}}_{\mathrm{Fe}} = 296$ eV (increases by 3.6%) and $I^{\mathrm{f}}_{\mathrm{Gd}} = 593$ eV (increase by 0.3%).

Let us assume that the increase in the mean excitation energy is caused by increase in binding energy of the n_x open shell electrons from $\hbar\omega^{\mathrm{p}}_x$ in paramagnetic phase to $\hbar\omega^{\mathrm{f}}_x = \beta\hbar\omega^{\mathrm{p}}_x$ in ferromagnetic phase. Then from Eq. (1) we find that

$$\beta = \exp\left(\frac{Z_a}{n_x}\ln\alpha\right) = \exp\left(-\gamma\frac{Z_a}{n_x}\ln\frac{E_{\mathrm{m}}}{I}\right). \tag{4}$$

If γ is measured with accuracy of $\mathrm{d}\gamma$ then $\mathrm{d}\beta = \beta\ln\beta\mathrm{d}\gamma$ and the experimental exchange energy can be determined with very high accuracy.

For 5.5 MeV α-particles in Fe ($3d^6$ electrons responsible for ferromagnetism, $\hbar\omega^{\mathrm{p}}_{3d} = 14.2$ eV) Eq. (4) with $\gamma = 0.015$ gives $\beta = 1.165$ and in consequence the additional binding energy in ferromagnetic state is $U_{\mathrm{Fe}} = 2.342$ eV. For 5.5 MeV α-particles in Gd ($4f^7$ electrons responsible for ferromagnetism, $\hbar\omega^{\mathrm{p}}_{4f} = 22.8$ eV) Eq. (4) with $\gamma = 0.002$ gives $\beta = 1.03$ and in consequence the additional binding energy in ferromagnetic state is $U_{\mathrm{Gd}} = 0.687$ eV. This energy is different for $3d^6$ and $4f^7$ electronic configurations.

Within the molecular field approach to description of ferromagnetism the exchange energy exerted from z nearest neighbours characterised by a spin vector S_j on each electron (of spin $s = 1/2$) belonging to atom located in the i-th lattice site is given by

$$U = -2Js\sum_{j}^{z} S_j \approx -2Jsz\langle S_j\rangle. \tag{5}$$

In this approach the exchange integral J is determined by the Curie temperature T_{C}. It is assumed that $z_{\mathrm{Fe}} = 8$, $z_{\mathrm{Gd}} = 12$, $z_{\mathrm{Ni}} = 8$. The average magnetisation $m = \langle S_j\rangle$ is given by the appropriate thermal average. It gives $J_{\mathrm{Fe}} \approx 2.4$ meV, $J_{\mathrm{Gd}} \approx 0.4$ meV, $J_{\mathrm{Ni}} \approx 1.3$ meV, and $J_{\mathrm{Co}} \approx 3$ meV. The addtional binding of the electron due to magnetisation at the lattice site is of the order of $U = mzJ$. This energy is $U_{\mathrm{Fe}} \approx 58$ meV, $U_{\mathrm{Gd}} \approx 17$ meV, $U_{\mathrm{Ni}} \approx 36$ meV, and $U_{\mathrm{Co}} \approx 83$ meV. This is much less than the additional binding energies U obtained from the energy loss measurement as calculated from the Bethe theory of ion stopping and also less than the thermal energy given in Table I, sufficient to destroy the ferromagnetism and cause the sample suffer the phase transition.

From Eqs. (1), (3) γ can be written also as

$$\gamma = -\sum_i n_i \ln\left(1 + \frac{U}{\hbar\omega_i}\right) / Z_a \ln\frac{E_{\mathrm{m}}}{I}, \tag{6}$$

TABLE I

From [14]: T_C — the thermal energy $k_b T_C$ [eV], ω_0 — the experimental plasmon energy [eV], E_F — the Fermi energy [eV], ρ — the density of quasi-free electrons [$e^-/Å^3$], ρ_i — density of interstitial charge [$e^-/Å^3$], n — the collective electrons per atom.

Elem.	T_C	ω_0	E_F	ρ	ρ_i	n
Fe	0.058	15.82	11.69	0.182	0.229	2.141
Co	0.121	17.85	13.74	0.231	0.228	2.575
Ni	0.054	20.44	16.46	0.304	0.232	3.325
Gd	0.025	10.45	6.72	0.079	0.098	2.618

From Eq. (6) it is obvious that the relative difference in the stopping power increases as $1/\ln E$ with decrease in the projectile energy E. It is worthy noting that the Zeeman-like splitting of the atomic energy levels with the theory of Eq. (1) yield $\gamma \propto -n_i \ln(1 - U^2/\hbar^2\omega_i^2) > 0$ indicating that such treatment fails in explanation of the measured effect in which $\gamma < 0$.

Another possible explanation of the effect is the hypothesis due to Stoner [12, 13] that in materials where the exchange interaction is large or the density of states is high at the Fermi level, as in case of $3d$ and $4f$ bands, we find spontaneous magnetic ordering *additionally* binding electrons with parallel spins. Particularly important are the quasi-free conduction electrons which, due to high plasmon excitation probabilities and energies given in Table I [14], are dominant in the electronic energy loss process. Also, the calculated collective electron density given in Table I reveals that more than two outermost electrons from each atom contribute to the electron gas collective response. Such the additional binding energies for the conduction electrons which enable reproducing the experimental data as for γ of Eq. (6) are: $U_{Fe} = 8.43$ eV, $U_{Gd} = 0.86$ eV.

Let us assume [15] a shift of energy $U_\pm$ of a conduction electron with spin up and down $\pm$ be proportional to the average number of electrons with spin up and down $N_\pm$ (without external magnetic field H):

$$U_\pm = -I_S N_\pm,$$

where I_S is the Stoner exchange integral. If band shift energy U and the magnetisation density M are defined by

$$U = \frac{I_S}{2}(N_+ + N_-), \qquad M = \frac{\mu_B}{V}(N_+ - N_-), \tag{8}$$

then

$$N_\pm = \frac{1}{2}\int_{U_\pm}^{\infty} \mathrm{d}E f(E - U_\pm) D(E - U_\pm). \tag{9}$$

where $f(E)$ is the Fermi–Dirac function and $D(E)$ is the density of states and

$$U_{\pm} = \mp \frac{I_S MV}{2\mu_B} - U. \tag{10}$$

From the standard procedure of decoupling magnetisation (Eq. (10) to Eq. (9) and to Eq. (8)) we get the Stoner conditions for band ferromagnetism

$$I_S D(E_F) \approx 2, \tag{11}$$

which links I_S and the density of states at the Fermi level. When we approximate the real density of states by the free electrons density of states, $D(E) = 1.5NE^{0.5}/E_F^{1.5}$, we can estimate the shift energy U for conduction electrons due to band ferromagnetism as

$$U \approx E_F/1.5. \tag{12}$$

We get $U_{Fe} \approx 7.8$ eV, $U_{Co} \approx 9.2$ eV, $U_{Ni} \approx 11.0$ eV, and $U_{Gd} \approx 4.5$ eV.

TABLE II

$-\Delta/S_e$(p) of the present measurement and of [9] and calculated along Eq. (6) in % for 5.486 MeV α-particles. n is the number of collective electrons per atom.

Elem.	Exp.	n = Table I	$n = 1$	$n = 2$
Fe	1.5 [9]	1.5	0.75	1.39
Co		2.6	1.14	2.06
Ni	1.6(±0.5)	2.3	0.71	1.38
Gd	0.2 [9]	0.9	0.36	0.70

The expected decrease in the α-particles stopping power during ferro-paramagnetic phase transition at T_C, calculated from Eq. (6) with additional binding energy due to the conduction electrons given by Eq. (12) is presented in Table II.

3. Experiment

The experimental setup consists of a reaction chamber of the pressure 10^{-5} Torr and thin source of α-particles of 5.486 MeV initial energy from ^{241}Am of 1 mC activity. The α-particles are collimated to the aperture of 6 mm diameter by foil holder and directed onto commercially available self-supporting foils of the thickness of 0.89 mg/cm^2 (corresponding to about 1 μm) from Ni ($T_C = 631$ K, $\rho = 8.8955$ g/cm^3).

The initial and final foil thickness is determined by measuring mass of the foil and subsequently by α-particles energy loss measurements and stopping and ranges of ions in matter (SRIM) data [14]. Both methods give the same results within 5% accuracy. When necessary, the foils could be made thinner (up to the 0.5 μm) by time-controlled solution in HF acid. The surface structure and composition

checkout are done by means of rf-spectrometer. This method can determine the contamination of the surface within accuracy of 1%.

The energy spectra of α-particles transmitted through the samples are registered with the Tennelec semiconductor surface barrier detector with 6 mm diameter diaphragm and cooled to temperature 252 K to reach the FWHM = 12 keV energy resolution. The temperature of the detector is controlled with a thermocouple attached directly to the detector. It turns out that the stabilisation of the detector temperature at a low (room) temperature is crucial in this experiment. An uncontrolled increase in the detector temperature causes decrease in signal amplitude and apparent increase in the ion energy loss. The ion signal from the detector is processed through the spectrometric CAMAC system and 4196 channel SWAN analyser.

The target foil is heated electrically by a wolfram spiral and the temperature is controlled with a thermocouple attached directly to the foil. The accuracy for setting the temperature is 1 K.

4. Results and discussion

The reference energy loss of 5.5 MeV α-particles in Ni is about 379.6 keV/μm [14] which yields the projected range of 10.5 μm allowing for analysis of quantities that are target thickness dependent. The reference energy spectrum measured for 5.486 MeV α-particles without foil was used for calibration of the spectrometer.

The energy spectrum measured for 5.486 MeV α-particles from ^{241}Am with foil was used for the foil thickness determination. The foil thickness along the ion beam increases with temperature and this increase approximately proceeds with the bulk thermal expansion coefficient: $\alpha_{\mathrm{Ni}} = 1.33 \times 10^{-5}$. The foil thickness in ferromagnetic phase at $T_{\mathrm{f}} = 293$ K was found to be $\delta x_{\mathrm{f}} = 0.985 \pm 0.01$ μm with the use of the reference energy loss [14]. The foil thickness in paramagnetic phase at $T_{\mathrm{p}} = 700$ K cannot be determined reliably with the reference energy loss since we expect its dependence on temperature. We use rather $\delta x_{\mathrm{f}} \rho_{\mathrm{Ni}}$ and $\delta x_{\mathrm{p}} \rho_{\mathrm{Ni}}$ as a measure of foil thickens, since we can measure the former and calculate the latter from thermal expansion coefficient α. Instead of simple energy loss per unit path length $\Delta E/\Delta x$ we use the stopping power $S_{\mathrm{e}} = \Delta E/\rho\Delta x$ of Eq. (1) in which surface density $\rho\Delta x$ at a given temperature is reduced to the density at initial temperature by means of thermal expansion coefficient.

The quantity of interest in the present experiment is the difference $\Delta E = \delta E_{\mathrm{f}} - \delta E_{\mathrm{p}}$. δE_{f} is the most probable energy loss calculated from energy spectra of α-particles transmitted through Ni foil in ferromagnetic phase at T_{f}. It was found $\delta E_{\mathrm{f}} = 369 \pm 12$ keV. δE_{p} is the most probable energy loss calculated from energy spectra of α-particles transmitted through Ni foil in paramagnetic phase at T_{p}. We got $\delta E_{\mathrm{p}} = 375 \pm 12$ keV. After correction for thermal expansion of the sample it yields from Eq. (3) $\gamma = 0.016 \pm 0.005$. This is the main experimental result of the work.

In Table II we compare the present experimental result as for relative increase in the electronic stopping power γ of α-particles in Ni foil after ferro-paramagnetic phase transition with previous experimental results for Fe and Gd and with theoretical results based on the Bethe theory of stopping with the Stoner itinerant electrons binding energy. The numerical results significantly depend on number n of collective electrons per atom accepted for calculations.

Other models that were used to explain the effect by assuming additional binding energy in ferromagnetic phase of outer shell electrons and conduction electrons due to electron spin interaction with magnetic field of the domain failed in comparison with experiment. The energy turned out too small to explain the measured stopping power increase.

5. Conclusions

We measured the relative difference in the stopping power of MeV α-particles in Ni foil subjected to the ferro-paramagnetic phase transition at T_C. It was found that for Ni foil $\Delta S/S_e(T_0) = 1.6\%(0.5\%)$. The relative difference of the stopping power calculated within the Bethe model of the ion stopping yields depends on the parameters of collective electrons per atom and on the parameters of spontaneous magnetisation of conduction electrons. The result is in agreement with the previous measurement showing that for neighbouring Fe foil the relative difference of stopping power amounts to 1.5%.

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